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1	Cosmogenic Nuclide Systematics and the CRONUScale
2	Program
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Abstract 13

As cosmogenic nuclide applications continue to expand, the need for a 14 common basis for calculation becomes increasingly important. In order to 15 accurately compare between results from different nuclides, a single method 16 of calculation is necessary. Calculators exist in numerous forms with none 17 matching the needs of the CRONUS-Earth project to provide a simple and 18 consistent method to interpret data from most commonly used cosmogenic 19 nuclides. A new program written for this purpose, CRONUScalc, is presented 20 here. This unified code presents a method applicable to ¹⁰Be, ²⁶Al, ³⁶Cl, ³He, 21 and ¹⁴C, with ²¹Ne in testing. The base code predicts the concentration of 22 a sample at a particular depth for a particular time in the past, which can 23 be used for many applications. The multi-purpose code already includes 24 functions for calculating surface exposure age for a single sample or for a 25 depth profile containing multiple samples. The code is available under the 26 GNU General Public License agreement and can be downloaded and modified 27 to deal with specific atypical scenarios. 28

Keywords: cosmogenic nuclide, exposure age calculator, beryllium-10, 29

chlorine-36, aluminum-26, helium-3, carbon-14 30

31 1. Introduction

The CRONUS-Earth Project, funded by the U.S. National Science Foun-32 dation, is intended to improve many aspects of cosmogenic isotope use and 33 help create a consistent, accurate use of the technique within the commu-34 nity. It functioned in collaboration with CRONUS-EU, a similarly motivated 35 group in Europe funded by the European Commission. One important part 36 of that project is the creation of a code that can consistently perform nec-37 essary calculations for different scaling schemes and that is applicable to 38 many of the commonly used nuclides (¹⁰Be, ²⁶Al, ³⁶Cl, ³He, ¹⁴C). The pro-39 gram presented here, called CRONUScale, is a joint effort by the CRONUS 40 Project to incorporate all major contemporary advances in the understand-41 ing of cosmogenic-nuclide production and to treat all the commonly used 42 nuclides in an internally consistent fashion. This calculator directly incor-43 porates much of the format and function of the ${}^{26}\text{Al}/{}^{10}\text{Be}$ code by Balco 44 et al. (2008), but it extends the functionality beyond ${}^{26}\text{Al}/{}^{10}\text{Be}$ as well as 45 introducing other new features. The new CRONUScalc code keeps the orig-46 inal modular format, but has updated the code with functions to perform 47 production/accumulation calculations, calibrations, and surface and depth 48 profile exposure ages and erosion rates. 49

The base code of CRONUScalc predicts the cosmogenic nuclide concen-50 tration in a sample (of either finite or point thickness) at a given depth 51 at a particular time in the past. This function allows great versatility 52 in earth science applications. CRONUScalc can be used to predict con-53 centrations of a suite of nuclides for a variety of purposes. The code is 54 published under the GNU General Public License, version 2 terms. The 55 basic code can be modified to output many different parameters and can 56 be downloaded (https://bitbucket.org/cronusearth/cronus-calc) and 57 modified to suit a user's particular needs. The code repository is open to user 58 contributions, allowing for sharing and future growth of the program. The 59 version of the program described in this paper is marked online as Version 60 2.0 in the downloadable repository. 61

The code has been used to create two specific calculators to address the common need to calculate surface exposure ages from unknown samples. The two calculators that are included with the program are a single-sample surface exposure age calculator and a multi-sample depth-profile calculator. Additional functions in the code are designed to calibrate production rates, test alternative scaling frameworks, and determine erosion rates.

The fundamental theory and assumptions that have gone into the code are 68 described in this paper. This paper matches the CRONUScalc downloadable 69 version 2.0 available in the repository. There are significant new features in 70 CRONUScale as compared to previous calculators, including a more accurate 71 method of integration through time and depth, updated geomagnetic history, 72 newly produced calibration data sets, updated and calibrated muon produc-73 tion model, and the ability to calculate exposure ages for single samples at 74 depth or perform a calibration. 75

76 2. CRONUScalc Program Systematics

The complete set of equations and fundamental theory behind the code 77 can be found in Appendix A and will be useful for those new to the topic 78 or interested in particular details because Appendix A systematically doc-79 uments all the equations used in CRONUScalc, without need for the reader 80 to refer back to numerous prior publications. The sections contained in the 81 main body of the paper are designed to provide a general cosmogenic user 82 with a summary of the systematics and relevant publications, focusing on the 83 new research developments compared to the code described in Balco et al. 84 (2008). A standard reference that describes the equations for production 85 of cosmogenic nuclides is Gosse & Phillips (2001). The equations given in 86 that paper have provided the baseline for numerous applications of cosmo-87 genic nuclides. CRONUScalc is an implementation of equations from Gosse 88 & Phillips (2001) and from newer sources, as described in each section. 80

90 2.1. Production Equations

91 2.1.1. Spallation

Cosmogenic-nuclide production from spallation, defined for this purpose 92 as the interaction of a high-energy particle with a target nucleus producing 93 a cosmogenic nuclide as a product of the reaction, follows a well-established 94 exponential decrease with depth. At the surface, spallation is typically the 95 dominant production mechanism. All the nuclides discussed in this paper 96 are produced through at least one spallation pathway. The formula for the 97 instantaneous production rate from spallation $(P_{s,m})$ is (Gosse & Phillips, 98 2001; Schimmelpfennig et al., 2008): gc

$$P_{\rm s,m}(Z) = S_T \sum S_{el,s} P_{\rm m,k}(0) C_{\rm k} \exp\left(-\frac{Z}{\Lambda_{\rm f,e}}\right),\tag{1}$$

where $P_{m,k}$ is the modern sea-level, high-latitude production rate of species 100 m by spallation of element k at 2010 solar modulation (atoms g⁻¹ a⁻¹); S_T 101 is the topographic shielding factor (unitless); $S_{el,s}$ is the geographical scaling 102 factor for spallation reactions for the particular reaction of interest, which 103 varies temporally due to fluctuations in the geomagnetic field or solar mag-104 netic field (unitless); C_k is the concentration of the element k (atoms g⁻¹) 105 a^{-1} ; and $\Lambda_{f,e}$ is the effective attenuation length for the spallogenic reactions 106 (applies to fast neutrons and spallogenic protons) (g $\rm cm^{-2}$). The production 107 is summed for all target elements k that produce nuclide m to give the total 108 spallation production rate. 109

110 2.1.2. Epithermal and Thermal Neutrons

Low-energy cosmogenic nuclide production, including that from thermal 111 and epithermal neutrons, does not follow a simple exponential pattern with 112 depth due to the atmosphere-ground interface effects. Low-energy neutrons 113 produced in the upper ~ 50 cm of rock tend to diffuse upward out of the 114 rock and into the atmosphere, resulting in a reduction of the flux as the rock 115 surface is approached. Phillips et al. (2001) analytically solved the neutron-116 flux differential equation across the land/atmosphere interface to obtain the 117 appropriate production equations. 118

The production of cosmogenic nuclides via the low-energy pathway is 119 dependent on the neutron flux, which is, in turn, dependent on the composi-120 tion of the rock. In order to calculate production from this pathway using a 121 universal parameter, the value $P_f(0)$ is calibrated instead of any parameter 122 dependent on composition. The $P_f(0)$ parameter is the production rate of 123 epithermal neutrons from fast neutrons in the air just above the surface. This 124 parameter can be used in conjunction with the sample-specific composition 125 to calculate the applicable production rates. 126

Cosmogenic nuclide production via low-energy neutrons is implemented 127 as described in Gosse and Phillips (2001), except for a small modification 128 to the calculation of muon-induced neutrons (discussed in Section 2.1.3). In 129 several cases, small typos in Gosse and Phillips (2001) were also corrected 130 (see the appendix for the corrected equations). Low-energy production is 131 currently only implemented for ³⁶Cl, but could be modified by advanced 132 users to apply to other nuclides with low-energy production pathways, such 133 as ³He. 134

135 2.1.3. Muons

Although muons make up a large portion of the incoming cosmic-ray flux at the earth's surface, the low interaction rate means that they penetrate more deeply into the subsurface than neutrons. Muon contributions to total cosmogenic nuclide production are typically small at the surface, but become important at depth (Stone et al., 1998). Accurate calculation of muogenic production is thus important for sampling sites with large erosion rates or samples at depth.

Early calculators employed an exponential approximation for production 143 from muons reactions according to Stone et al. (1998). Balco et al. (2008) 144 implemented a newer model by Heisinger et al. (2002b,a) that calculates 145 the production from muon reactions using physical parameters determined 146 from laboratory irradiation experiments. Braucher et al. (2003) used a deep 147 core to provide evidence that the parameters specified by Heisinger et al. 148 (2002a,b) overestimated actual ¹⁰Be production by fast muons by approxi-149 mately a factor of two. This was supported by additional profile data mea-150 sured by Braucher et al. (2011) and by Kim & Englert (2004), as well as 151 reanalysis of previously published deep profile data by Braucher et al. (2013). 152 Rather than using parameters estimated from laboratory muon irradia-153 tions, the CRONUS-Earth Project has adopted values calibrated from nuclide-154 concentration profiles at carefully selected sites (Fig 1 in Phillips et al., 2015; 155 Marrero, 2012; Borchers et al., 2015). A 30-m deep quartzite core from 156 Antarctica has allowed the calibration of muon production parameters for 157 ²⁶Al and ¹⁰Be (Fig 1 in Phillips et al., 2015). For ³⁶Cl, previously published 158 data sets (Evans, 2001; Stone et al., 1998) collected from quarry profiles were 159 used to reparameterize the muon model for cosmogenic ³⁶Cl production from 160 muons on Ca and K (Marrero, 2012). A CRONUS-Earth depth profile was 161 not measured for ${}^{14}C$, and ${}^{14}C$ profiles measured by other investigators (Kim 162 et al., 2007; Lupker et al., 2013, 2015) have shown problems with distinguish-163 ing slow-muon production from production by muogenic neutrons, therefore 164 at present CRONUScalc uses the values from Heisinger et al. (2002a.b) for 165

¹⁴C. This may result in less accuracy for the calculation of muogenic ¹⁴C
¹⁶⁷ production than for other nuclides, but ongoing investigations (Lupker et al.,
¹⁶⁸ 2013, 2015) may yield improved parameter values in the near future.

¹⁶⁹ Nuclide production by fast muon reactions $(P_{\mu,fast})$ is described by Equa-¹⁷⁰ tion 2 (Heisinger et al., 2002b). The parameter σ_0 was selected as the cali-¹⁷¹ bration parameter for the production of nuclides by muon reactions, as discussed in (Fig 1 in Phillips et al., 2015). This was mainly because it is the only nuclide-dependent parameter in the fast production equation. However, direct calibration of the σ_0 parameter also eliminates any dependence on the accuracy of the conversion from σ_{190} (the parameter measured in the laboratory irradiation experiments) to σ_0 (see Equation B.27, from Equation 14 in Heisinger et al. (2002b)).

$$P_{\mu,fast} = S_T \phi_{\mu,total}(\mathbf{Z})\beta(\mathbf{Z})(\bar{\mathbf{E}}(\mathbf{Z}))^{\alpha}\sigma_0 \mathbf{N}_{t,i}$$
(2)

Where the factor $\beta(Z)$ is a function of the mean total muon energy and 178 is shown in Equation B.25 and E is defined as the mean muon energy at a 179 given depth Z and is shown in Equation B.26. $\phi_{\mu,total}$ is the total muon flux 180 at the site, as calculated by the same equations used to calculate the fluxes 181 for the Lifton et al. (2014) scaling framework. $N_{t,i}$ is the number density of 182 the atoms in the target element (in units of at/g). This value is a constant 183 for each nuclide unless the composition of the target changes, as it does for 184 ³⁶Cl. α is an energy-dependent coefficient that parameterizes the energy 185 dependence of the cross-section (σ) on muon energy. Experimental results 186 permit values for α between 0.75 and >1.0 (Heisinger et al., 2002b) so the 187 CRONUS-Earth Project chose a value of $\alpha = 1.0$. By assuming that α equals 188 one, β will also be equal to one. 189

Nuclide production by slow negative muon capture $(P_{\mu-})$ is described by 190 Equation 3, originally from Charalambus (1971) and discussed in detail for 191 ³⁶Cl by Stone et al. (1998). The production rate depends on the stopping rate 192 of negative muons $(\phi_{\mu-})$ as well as the nuclide-dependent factors $(f_{i,C}, f_{i,D})$ 193 f_i^*). $\phi_{\mu-}$ is derived from the muon flux calculated by the same equations used 194 in Lifton et al. (2014). $f_{i,D}$ is the fraction of muons stopped by element k and 195 absorbed by the nucleus before decay of the muon. $f_{i,C}$, the compound factor, 196 represents the fraction of the muons that are captured by a target element 197 (as opposed to the other elements present) within the bulk rock. The formula 198 for the compound factor (Equation E.40) is taken from Charalambus (1971) 199 and the values are consistent with those used by Heisinger et al. (2002a). 200

$$P_{\mu-} = S_T \phi_{\mu-}(Z) f_{i,C} f_{i,D} f_i^* \tag{3}$$

The remaining parameter, f_i^* , the particle emission channel probability, is the probability that the excited nucleus of the target atom will emit the proper particle to result in transformation to the nuclide of interest. Heisinger et al. (2002a) experimentally determined f_i^* values for the production of ²⁶Al (from Si), ¹⁰Be and ¹⁴C (from O), and ³⁶Cl (from K and Ca), but these values tend to overestimate nuclide concentrations measured in depth profiles in the same fashion as for fast muon production (Fig 1 in Phillips et al., 2015; Braucher et al., 2011). The production parameters for slow negative muon capture, f_i^* , were calibrated by fitting to the measured CRONUS-Earth profile for for ¹⁰Be and ²⁶Al and by fitting to previously published profiles for ³⁶Cl, as previously discussed.

The muon-capture reactions can release neutrons that later participate 212 in neutron-capture reactions (muon-induced neutrons). Instead of assuming 213 an exponential decrease in muon production with depth to calculate this 214 flux, as most previous calculators do, the new muon module described above 215 is used to calculate production with depth. The muon code calculates the 216 negative muon stopping rate $(\phi_{\mu-}(Z))$ and total muon flux $(\phi_{\mu f}(Z))$ terms 217 at a given depth for the calculation of the production rate of muon-induced 218 neutrons (Equation 4). This muon-induced neutron production rate is used 219 in the thermal and epithermal neutron flux equations (Equations B.30 and 220 B.34), which are ultimately used to determine the production rate via the 221 low-energy neutron pathway. 222

$$P_{n,\mu}(Z) = Y_s \phi_{\mu-}(Z) + 5.8 \times 10^{-6} \phi_{\mu f}(Z), \qquad (4)$$

where Y_s is the average neutron yield per stopped negative muon (Fabryka-Martin, 1988).

225 2.1.4. Radiogenic Production

Radiogenic production in this context refers to the generation of low-226 energy neutrons by reactions related to the radioactive decay or spontaneous 227 fission of U or Th, and the subsequent absorption of those neutrons to pro-228 duce nuclides of interest, principally ³⁶Cl. Although ³He is also produced 229 from Li in this manner (Lal, 1987; Dunai et al., 2007), only ³⁶Cl radiogenic 230 subtraction is incorporated into the program and the discussion below focuses 231 on ³⁶Cl for that reason. The radiogenic low-energy neutron flux is assumed to 232 be in equilibrium with the concentrations of uranium (U) and thorium (Th) 233 in the rock. This component is quantified using measured concentrations 234 of U and Th and the method described in Fabryka-Martin (1988), which is 235 based on the formulations developed by Feige et al. (1968). These equations 236 are shown in Appendix B.5. 237

The quantification of the bulk rock properties requires fundamental nuclear properties for each element. The original table of these properties presented in Fabryka-Martin (1988) has now been corrected and updated using
the information in Mughabghab (2006) and Schimmelpfennig et al. (2009).
A table of nuclear properties for all the elements considered in CRONUScalc
is shown in Table 1.

In most studies, the rocks (not the exposure age) are sufficiently old that 244 the assumption of equilibrium is reasonable. Even for younger features, such 245 as a newly-erupted basalt, the assumption of equilibrium is reasonable due to 246 equilibration with the uranium and thorium concentrations prior to eruption, 247 so long as the magma did not undergo differentiation or mixing that signifi-248 cantly changed its U and Th contents less that ~ 0.5 Ma prior to sampling. 240 The alternative is to allow production from radiogenic sources to begin at 250 the exposure age (or any rock formation age). This difference in assumption 251 is small for most ³⁶Cl samples and no data set currently exists to differen-252 tiate between the two assumptions. This assumption is only significant for 253 young samples with high chlorine contents (> 50 % production from Cl) or 254 significant U or Th. 255

256 2.2. Calibration Technique

The production rates incorporated into the CRONUScalc code are the 257 results from CRONUS-Earth Project calibrations. The calibration data set 258 compiled by CRONUS-Earth includes a large number of sites representing a 259 large range of latitudes, longitudes, and elevations. Ideal sites should have 260 little uncertainty in the sample parameters (e.g. erosion rate, elevation, 261 shielding, independent age constraints, etc.). The sites were divided into 262 two categories based on the quality of the site, with the sites that came clos-263 est to fitting the criteria above placed into the 'primary calibration data set'. 264 Additional sites with independent age constraints, but not meeting the strict 265 criteria for primary sites, were placed into the 'secondary data set'. These 266 samples are independent of the primary calibration data set and therefore 267 can be used to assess the final calibrated parameters. 268

The calibration method, data set details including references, and the spallation calibration results for all nuclides are discussed in detail in Borchers et al. (2015). Additionally, some details of the ¹⁰Be and ²⁶Al muon calibration can be found in Phillips et al. (2015) and the full chlorine-36 calibration can be found in Marrero (2012); Marrero et al. (2015).

EI.	A_i	ξ_i	$\sigma_{sc,i}$	$\sigma_{th,i}$	$I_{a,i}$	S_i	$Y^u_{n,i}$	$Y^{th}_{n,i}$	K_m	St.	#	Ь
	[g/mol]	[-]	[q]	[b]	$[1 \times 10^{-24}]$	[-]				_	_	_
0	16	0.120004104	3.761	0.00019	0.0002693	539	0.23	0.079	0	0	∞	1.00
Н	1.01	1	20.49	0.3326	0	1542	0	0	0	0.5		0.00
C	12.01	0.157760474	4.74	0.0035	0.0018	573	0.45	0.18	13.691	2	9	0.36
Na	22.99	0.084543589	3.038	0.517	0.311	454	14.5	6.8	19.42	0.5	11	1.00
Mg	24.31	0.08009077	3.414	0.0666	0.038	463	5.8	2.6	14.94	-	12	0.93
Al	26.98	0.072337427	1.413	0.231	0.17	449	5.1	2.6	11.812	1.5	13	0.76
Si	28.09	0.069559975	2.044	0.171	0.082	455	0.69	0.335	10.01	5	14	0.84
Ч	30.97	0.063210393	3.134	0.165	0.079	444	0	0	8.48	2.5	15	1.04
Х	39.1	0.050295528	2.04	2.1	1	432	0.45	0.305	12.78	0.5	19	1.54
Ca	40.08	0.049086179	2.93	0.43	0.233	436	0	0	10.73	, _ i	20	1.90
: Li	47.87	0.041208508	4.09	6.41	3.1	367	0	0	7.53	2	22	2.66
Mn	54.94	0.035968204	2.06	13.36	13.4	351	0	0	8.486		25	2.73
Fe	55.85	0.035390922	11.35	2.56	1.36	353	0.19	0.205	7.54	1.5	26	3.28
C	35.45	0.055371497	15.8	33.14	13.83	420	0	0	16.98	0	17	1.32
В	10.81	0.174236264	4.27	767	343	537	62.3	19.2	55.68	0	ы	0.25
Sm	150.36	0.013242694	38	9640	1400	0	0	0	4.004	0	62	4.4
IJ	157.25	0.012664667	172	41560	390	0	0	0	3.828	0	64	5.8
Ŋ	238.03	0.008378873	9.08	2.68	277	0	0	0	2.529	0	92	4.7
Th	232.04	0.008594581	13.55	7.34	83.3	0	0	0	2.594	0	90	3.0
Li	6.9	0.264	0.95	70.5	0.0	548	21.1	9.6	86.731	0.0	3	0.18
Cr	52.0	0.038	3.38	3.1	1.6	0.0	0.0	0.0	11.578	0.0	24	2.98
		-										

Note: 1 barn $(b)=1\times10^{-24}$ cm². The columns correspond to the following parameters: A_i - Atomic weight of integral for element i; S_i - Mass stopping power of element i for alpha particles of a given energy; $Y_{n,i}^u$ neutron yield of element i per ppm U in radioequilibrium; $Y_{n,i}^{th}$ - Neutron yield of element i per ppm Th in radioequilibrium; K_m - 602/atomic weight of sample, used to convert to at/g from ppm; St. - Stoichiometric Table 1: Table of constants used for elemental parameters. The table was originally taken out of Fabryka-Martin (1988). It has been updated using Mughabghab (2006). El is the element; St is the stoichiometry. element; ξ_i - Average log decrement of energy per neutron collision with element i; $\sigma_{sc,i}$ - Neutron scattering cross-section of element i; $\sigma_{th,i}$ - Thermal neutron absorption cross-section of element i; $I_{a,i}$ - Dilute resonance tatio of oxide (oxygen to element i); # - Atomic number; **P** - Average capture probability relative to oxygen.

274 2.3. Production Rate Scaling

Cosmogenic nuclide scaling applies the physics governing the modulation 275 of the cosmic-ray flux by atmospheric mass and the terrestrial and solar 276 magnetic fields to provide production rates as a function of location and ex-277 posure time. Numerous scaling frameworks have been proposed in order to 278 correct for latitude, elevation, atmospheric pressure anomalies, dipole and 279 non-dipole geomagnetic field changes, and solar modulation. CRONUScalc 280 implements seven scaling frameworks, as summarized in Table 2. This in-281 cludes the original model by Lal (1991), further developed by Stone (2000) 282 (referred to as St), that was based on cosmic-ray emulsion data and some 283 neutron monitor data. A version of the Lal/Stone scaling accounting for ge-284 omagnetic field variations is also included (Lm). CRONUScalc includes the 285 neutron-monitor-based models of Dunai (Du) (Dunai, 2000, 2001a), Lifton 286 (Li) (Lifton et al., 2005), and Desilets (De) (Desilets & Zreda, 2003; Desilets 287 et al., 2006b). Finally, CRONUScalc implements two new models based on 288 Lifton et al. (2014). The Lifton-Sato-Dunai (LSD, denoted SF herein) scaling 289 framework (Lifton et al., 2014) does not rely on empirical fitting to a set of 290 measured data points, but instead is based on analytical fits to physics-based 291 modeling, and has been tested against measured atmospheric secondary nu-292 cleon and muon energy spectra. 293

The energy spectrum of the cosmic-ray flux produced by the LSD frame-294 work provides information that is available neither from the traditional Lal 295 (St) scaling, nor neutron-monitor based scaling (Du, Li, De). This, in turn, 296 allows the production rates of cosmogenic nuclides to be calculated by inte-297 grating the energy-dependent excitation function with the calculated particle 298 energy spectrum. This is termed "nuclide-dependent scaling" because it re-299 sults in a separate scaling factor for each nuclide, rather than a single scaling 300 factor applicable to all nuclides, as the previous methods did. In the case of 301 ³⁶Cl, for example, this results in six nuclide-dependent scaling factors, includ-302 ing four high-energy scaling factors to scale the spallation reactions (K, Ca, 303 Ti, and Fe) and one generalized spallation reaction to scale the low-energy 304 reactions. Due to the derivation from the high-energy flux, the low-energy 305 pathways cannot be scaled using the incoming low-energy neutron flux. (Note 306 that since ³⁶Cl is commonly measured in rocks of varying compositions, each 307 reaction has to be individually weighted by the concentration of the target 308 element in the rock, unlike nuclides such as ¹⁰Be or ²⁶Al that are commonly 309 measured in minerals of constant composition, such as quartz). In all of the 310 other scaling frameworks, the nuclide-dependent scaling factors still appear in 311

the code, but all are set equal to the single scaling factor produced by the se-312 lected scaling framework. This nuclide-dependent scaling framework (LSDn, 313 denoted SA herein) is the default on the online interface due to its favorable 314 calibration results (see Borchers et al. 2015) and the possibility for future 315 incorporation of updates to the excitation functions and other physics-based 316 research. For all the details of the scaling frameworks themselves, please see 317 the original papers (cited in Table 2); for details of implementation, please 318 see the description in Balco et al. (2008) and Lifton et al. (2014). 319

In CRONUScale, the geomagnetic history is consistent across all scaling 320 frameworks, even though each model uses the history in a slightly differ-321 ent manner. This information is available in the code and can be modified 322 directly if a different geomagnetic history is necessary. References for the 323 currently implemented geomagnetic history can be found in Table 3, with 324 relevant details in Lifton et al. (2014). The calculator is flexible, allowing 325 knowledgeable users to modify the code to incorporate alternative geomag-326 netic models. 327

Although the various paleomagnetic reconstructions are in broad agree-328 ment, there are considerable differences for certain time intervals, indicating 329 some uncertainty with regard to the accuracy of each reconstruction. In 330 addition, each reconstruction contains internal uncertainties stemming from 331 uncertainties in the measurements and chronology. We do not attempt to 332 propagate these uncertainties into the CRONUScalc ages. This is partly due 333 to the difficulty of quantifying the unknown errors in each reconstruction 334 (for example, the assumed constant geomagnetic field of the St model is also 335 clearly in error, but how does one quantify the magnitude and propagate it 336 into the resultant age?). When considering the entire CRONUS-Earth sec-337 ondary calibration data set (Borchers et al., 2015), the difference between 338 the average normalized Lal-Stone (St; constant geomagnetic field) ages and 339 the Lal (1991) (Lm; time-varying field) ages is only 0.4%, and increases to 340 1.8% for the same comparison at the high-elevation, low-latitude Breque, 341 Peru, site. However, due to the approximately sinusoidal dipole variation 342 over the exposure time of a significant number of the calibration sites, these 343 comparisons may still underestimate the differences when sample exposure is 344 integrated over longer time spans in which the time-integrated effects might 345 not cancel (<ca. 100 ka). Considering the difficulty of quantifying this un-346 certainty and propagating it, and the minimal effect the effort would have 347 on age uncertainties for a majority of samples, we have neglected it. We 348 acknowledge that for older or younger samples especially at low latitude, the 349

Abbr.	Reference	Description
St	Lal (1991) ; Stone	Time-independent (constant production rate). Based on two
	(2000)	different types of neutron reaction counting methods (photo-
		graphic emulsions and neutron monitors). Scaling factor inputs
		are geographic latitude and atmospheric pressure, based on the
		Stone (2000) modification.
Lm	Lal (1991); Nishi-	Time-dependent version of St based on time-variation in the
	izumi et al. (1989)	dipole magnetic field intensity, as formulated by Nishiizumi
		et al. (1989).
Li	Lifton et al. $(2005,$	Time-dependent model based on neutron monitor measure-
	2008)	ments and incorporating dipole and non-dipole magnetic field
	,	fluctuations and solar modulation. The scaling factor is based
		on actual atmospheric pressure, solar modulation, and a cutoff
		rigidity calculated using trajectory tracing.
Du	Dunai (2000,	Time-dependent model based on neutron monitor measure-
	2001a,b)	ments and incorporating dipole and non-dipole magnetic field
		fluctuations. The scaling factor is based on an analytically cal-
		culated cutoff rigidity and atmospheric pressure. A long-term
		mean for solar modulation is used in this model.
De	Desilets et al.	Time-dependent model based on neutron monitor measure-
	(2006b); Desilets	ments and incorporating dipole and non-dipole magnetic field
	& Zreda (2003)	fluctuations. The scaling factor is based on a cutoff rigidity
		calculated using trajectory tracing and the actual atmospheric
		pressure.
LSD	Lifton et al.	Time-dependent model based on equations from a nuclear
(Sf)	(2014)	physics model and incorporating dipole and non-dipole mag-
		netic field fluctuations and solar modulation. The scaling factor
		is based on actual atmospheric pressure, solar modulation, and
		a cutoff rigidity calculated using trajectory tracing. The total
		flux (protons+neutrons) is used to scale all reactions.
LSDn	Lifton et al.	Time-dependent model based on equations from a nuclear
(Sa)	(2014)	physics model and incorporating dipole and non-dipole mag-
. ,		netic field fluctuations and solar modulation. The scaling
		factor is based on actual atmospheric pressure, solar modu-
		lation, and a cutoff rigidity calculated using trajectory trac-
		ing. Nuclide-dependent scaling is implemented by incorporating
		cross-sections for the different reactions.

Table 2: Table of scaling frameworks, including abbreviations, original references, and a brief description of each model.

Time period (kyr)	Model name	Reference
3-7ka	CALS7k	Korte & Constable (2005)
7-18ka	GLOPIS-75	Laj et al. (2004)
18ka-2Ma	PADM2M	Ziegler et al. (2011)

Table 3: Geomagnetic history used in all the scaling frameworks.

effect might be larger and we hope that quantification will be addressed in
future research. The geomagnetic models used may also include systematic
spatial or temporal biases that may be resolved with further research.

While CRONUScalc offers seven different spallation scaling frameworks, it uses only one muon scaling model. Muons were not natively scaled in any of the original models, so we use the energy-dependent Lifton et al. (2014) muon model to scale the muon flux.

A new input to the code is the "year collected" that allows the scaling 357 frameworks to more precisely account for the time of exposure. In previous 358 calibrations, the independent age constraints have been listed as "before 350 present." For radiocarbon analyses, this is actually "years before 1950." 360 The calibrations have been updated accordingly and now list independent 361 ages as "years before 2010" and the exposure age calculation routines in 362 CRONUScale now integrate production through the collection year. For 363 most samples, this is not a critical change, but with the advent of very high-364 precision cosmogenic ages on very young samples (e.g. Schaefer et al. 2009), it 365 will eliminate the need for ad-hoc corrections for sample date. This capability 366 will only become more important through time. The sample is assumed to 367 have been processed shortly after collection or to have been stored where 368 continued production and decay would be insignificant. 369

370 2.4. Attenuation Length

The apparent attenuation length, the attenuation length with respect to a flat surface with no topographic shielding, quantifies the depth distribution of the production by neutron spallation. When the apparent attenuation length is adjusted to account for the dip of the sample surface and any topographic shielding, the result is the effective attenuation length, $\Lambda_{f,e}$. The effective attenuation length is the parameter that should be used in calculations pertaining to production from a particular sample.

The effective attenuation length is an input parameter for each sample. The web-based user interface automatically calculates the effective attenua-

Depth/Cut. Rig.	1100	1000	900	800	700	600	500	400
0	151	152	153	154	155	158	163	172
4	152	152	154	156	159	164	171	185
8	156	159	161	164	168	175	185	204
12	162	165	168	171	176	184	196	218
16	168	170	172	177	182	191	204	228
20	169	171	174	179	184	192	206	231

Table 4: The table used for the interpolation of attenuation lengths based on the given atmospheric depth (top row, in units of g/cm^2) and cutoff rigidity (first column, in units of GV). Values based on those obtained using the spreadsheet that accompanied Sato et al. (2008), but include the additional 11.1% correction discussed in the text.

tion length if it is not provided. For use in the raw code where attenuation 380 length is not automatically calculated, there is a function (attenuationlengthnormal.m) 381 to calculate the parameter at a given location (latitude, longitude, elevation) 382 and pressure of a sample. The attenuation-length model in CRONUScalc is 383 based on atmospheric attenuation lengths calculated from the PARMA model 384 of Sato et al. (2008), which is an analytical model for estimating cosmic-ray 385 spectra in the atmosphere. The attenuation lengths are interpolated from 386 the values given in Table 4 using the vertical cutoff rigidity and atmospheric 387 depth of the site. 388

The interpolated attenuation lengths have been adjusted upward by 11.1% to account for systematic differences between atmospheric and lithospheric attenuation. This adjustment is based on the approximate relation:

$$\frac{\lambda_{f,rock}}{\lambda_{f,atm}} = \left(\frac{A_{rock}}{A_{atm}}\right)^{1/3} \tag{5}$$

where A_{rock} and A_{atm} are the average atomic weights of the rock and atmosphere, respectively. The average atomic weight of the atmosphere was taken to be 14.68 g mole⁻¹ and that of average rock 20.40 g mole⁻¹. (We note that any changes in the chemical composition of the atmosphere over the time scale of interest for application of cosmogenic-nuclide surface chronology are too small to affect the attenuation length.) This relation is derived from the following simple considerations.

The macroscopic cross-sectional area of the atoms in a gram of matter scales as the product of the number of atoms of each element in the volume

multiplied by the average cross-sectional area of the atoms. Assuming that 401 atomic nuclei are spheres of constant density, and that each nucleus is made 402 up of spheres of uniform mass and diameter (protons and neutrons), the 403 radius of each atom will scale by the atomic weight to the one-third power 404 (Friedlander et al., 1981). The cross-sectional area will then scale as the 405 square of the radius. The product of these is the average atomic cross-406 section to the two-thirds power. The number of atoms per gram will scale 407 by Avogadro's number divided by the average atomic weight. Therefore, 408 the macroscopic cross-sectional area of substance i (rock or atmosphere), per 400 gram, will scale as follows: 410

$$Area_i \propto \frac{N_A A_i^{2/3}}{A_i} = \frac{N_A}{A_i^{1/3}} \tag{6}$$

where N_A is Avogadro's number. The apparent attenuation length is the inverse of the macroscopic area per gram. When the ratios of the attenuation lengths for rock and atmosphere are taken, Equation 5 results. (John Stone, personal communication, 14 February 2012).

The attenuation length will also vary as a function of lithology. The 415 average atomic weight of common lithologies varies from 19.8 g mole⁻¹ for 416 quartzite to 21.5 g mole^{-1} for ultramafic rocks. However, the maximum 417 difference between the attenuation length calculated for any lithology and 418 that for the mean atomic weight (averaged over all lithologies) is < 2 %. 419 This is insignificant and hence CRONUScalc does not compute attenuation 420 lengths on a lithology-specific basis. There is some modeling evidence to 421 suggest that nuclide-dependent attenuation lengths may be needed in the 422 future (Argento et al., 2014), something the calculator is already equipped 423 to accommodate because attenuation length is a sample input. 424

Figure 1 shows the dependence of the modeled effective attenuation length on elevation and latitude. For samples with large topographic shielding corrections, these values should be adjusted to account for horizon obstructions.

The final attenuation lengths overlap with the range of values presented by Dunai (2000) from 121 to >170 g/cm², with the range for most samples falling between 150-190 g/cm² (see Figure 1). Although the results are in the same range as commonly assumed values of 160-170 g cm⁻² (Gosse & Phillips, 2001; Dunai, 2010), a significant difference in age between the use of the two different attenuation lengths (CRONUS-predicted value vs 170 g cm⁻²) is possible for samples with high erosion rate, very long exposure times, or at

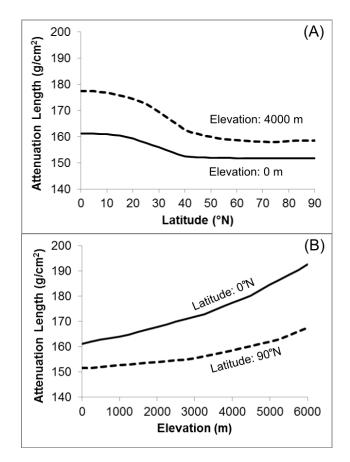


Figure 1: The change in effective attenuation length for a flat surface at various elevations (A), shown for both zero and 90°N latitude, and effective attenuation length at various latitudes (B), shown for elevations of 0 and 4000 m.

significant depth. In order to test the magnitude of this effect, we predicted 435 ¹⁰Be concentrations using the CRONUScalc modeled attenuation length of 436 153 g/cm^2 (corresponding to 40°N at sea level) for artificial samples for a 437 range of ages and erosion rates. The resulting ages were then determined 438 using an attenuation length of 170 g/cm². The results (Figure 2, B.6) show 439 that the attenuation length change from 153 g/cm^2 to 170 g/cm^2 results in 440 up to a 2% difference on the 20 ka age for erosion rates up to 10 mm/kyr. 441 The difference is considerably larger for faster eroding samples with longer 442 exposure times, or alternately samples at depth, with differences of almost 443 50 % for 300 ka exposure times with erosion rates of 10 mm/kyr. However, 444 for most surface samples with modest or low erosion rates, this difference is 445 relatively small. When depth profiles are available, the attenuation length 446 should still be one of the fitted parameters to ensure the most accurate value 447 (e.g. Braucher et al. 2013). Use of the surface shielding calculator to estimate 448 attenuation lengths for depth profiles or very high erosion rates should be 449 approached with caution. 450

Given the physics-based derivation of the new attenuation length model, 451 the new attenuation lengths are implemented as the default in the online 452 calculator interface. However, as this parameter is an input for each sample, 453 the choice of attenuation length is ultimately left to the user. The final model 454 for attenuation length is included as its own function in the code repository 455 (see the Function Appendix in the supplementary materials). A separate 456 shielding calculator has been developed to simplify the calculation of the 457 topographic shielding factor as well as the appropriate effective attenuation 458 length. The attenuation length model accounts for the latitude and elevation 459 of a sample. If shielding information is included, the effective attenuation 460 length is modified accordingly. A link to the shielding calculator is available 461 on the calculators homepage (http://web1.ittc.ku.edu:8888/. 462

463 2.5. Accumulation

Instantaneous production rates, such as those described above, must be integrated in both time and thickness in order to calculate the production in a real sample. In many common solutions, this is done analytically or with additional multiplicative factors to account for the finite thickness of a sample. In the CRONUScalc program, this integration is done numerically, which is more accurate for depth integrations and makes it simple to incorporate time-dependent scaling and varying production rates.

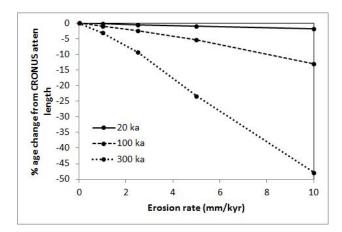


Figure 2: Change in ages due to change in attenuation length from the original CRONUS-calculated value (153 g/cm^2) to 170 g/cm^2 , compared over a range of erosion rates (0, 1, 2.5, 5, and 10 mm/kyr) and three different exposure ages (20, 100, and 300 ka).

For a given age, an appropriate time step, Δt , is found such that the erosion rate is small for that period and the changes in time-dependent production rates are not too large. Although larger time steps may be appropriate for some samples or scaling methods, the time step default is set to 100 years in order to maintain time-step errors below 0.4% for all sample types, as calculated assuming the correct age is the age produced using a time step 477 of 1 year.

The sample begins at depth and the cosmogenic inventory within the sample is tracked during its migration to the surface. For each time step, the current scaling factors and production rates are found for the sample. The inventory accounts for the decay of all previously produced nuclide inventory (the first term in Equation 7) as well as the production and decay of the nuclide during the current time step (second term in Equation 7).

$$N_{tot} = N_{prev}e^{-\lambda\Delta t} + P_{tot} * f_{decay} \tag{7}$$

where N_{tot} is the total inventory in the sample up to the current time step; N_{prev} is the inventory from all previous time steps; λ is the decay constant for the nuclide; P_{tot} is the instantaneous production rate of the nuclide from all mechanisms; and f_{decay} is the decay factor (Equation 8) that accounts for the fact that a small amount of the nuclide produced at the beginning of the time period will have decayed by the end of the period.

$$f_{decay} = \frac{1 - e^{-\lambda \Delta t}}{\lambda} \tag{8}$$

After the nuclide is produced, the samples are "eroded" by updating the depths for the sample by using the erosion rate and the time step, as shown in Equation 9.

$$D_{new} = D_{previous} - \Delta t * \epsilon \tag{9}$$

where D_{new} is the new depth, D_{prev} is the previous sample depth, and ϵ is the erosion rate of the surface.

These steps are repeated from t=(year sampled)-age, with the sample at depth, until t is equal to the year sampled, when the sample reaches the surface. The inventory is then returned by the module and used in any number of other functions, such as surface or depth-profile calculators, or simply returned to the user.

⁵⁰⁰ 3. Considerations Specific to Individual Cosmogenic Nuclides

While the in-situ cosmogenic nuclides share many similarities in terms of production pathways, they are each produced by a different combination of pathways and there are other individual issues that need to be addressed. In the following sections, the specific pathways for production, uncertainties, and other issues are documented for each of the major four code modules representing five nuclides.

Each nuclide is produced by one or more of the pathways discussed in the previous section. The most common reactions are shown in Table 5 for common nuclides and their common target minerals.

Inputs required by CRONUScalc vary, but the inputs common to all nu-510 clides include location (latitude, longitude, elevation/pressure), sample pa-511 rameters (thickness, bulk density, depth, and attenuation length), site pa-512 rameters (topographic shielding, erosion rate, year collected), and chemical 513 parameters (cosmogenic nuclide concentration). Additional inputs are neces-514 sary for some nuclides to account for production through low-energy neutron 515 pathways (composition, water content). For all nuclides, the required cosmo-516 genic nuclide concentration is assumed to incorporate any blank corrections, 517 as these vary from lab to lab and therefore cannot be standardized to be 518 included in the program. 519

Nuclide	Reaction
³ He	Spallation: O, Mg, Si, Ca, Fe, Al
	$^{6}\text{Li}(n,\alpha)^{3}\text{He}$
¹⁰ Be	$^{16}O(n,4p3n)^{10}Be$
	${}^{28}{\rm Si}({\rm n,x}){}^{10}{\rm Be}$
	$^{16}O(\mu^{-},\alpha pn)^{10}Be$
	${}^{28}\text{Si}(\mu^-,\mathbf{x}){}^{10}\text{Be}$
¹⁴ C	$^{16}O(\mu^{-},2p)^{14}C$
	$^{16}O(n,2pn)^{14}C$
	$^{17}O(n,\alpha)^{14}C$
	${}^{28}{ m Si}({ m n,x}){}^{14}{ m C}$
²⁶ Al	$^{28}{ m Si}({ m n},2{ m pn})^{26}{ m Al}$
	$^{28}\text{Si}(\mu^-,2\text{n})^{26}\text{Al}$
³⁶ Cl	$^{40}Ca(n,2n3p)^{36}Cl$
	39 K(μ^{-} ,p2n) ³⁶ Cl
	$^{40}\mathrm{Ca}(\mu^-,lpha)^{36}\mathrm{Cl}$
	$^{35}\mathrm{Cl}(\mathrm{n},\gamma)^{36}\mathrm{Cl}$
	39 K(n, α) ³⁶ Cl
	$Fe(n,x)^{36}Cl$
	$Ti(n,x)^{36}Cl$

Table 5: Common reactions producing in-situ terrestrial cosmogenic nuclides (Gosse & Phillips, 2001). The symbolism is as follows for the X(a,b)Y reaction: X is the target element, a is the particle interacting with the target, b is emitted during the reaction, and Y is the final nuclide product. "x" in the second place inside the parentheses indicates a reaction for which several different combinations of emitted particles are possible. Reactions are only shown for the pathways discussed in the paper. For beryllium, aluminum, and carbon, only those reactions possible in a quartz target mineral are shown.

520 3.1. Aluminum-26 and Beryllium-10

¹⁰Be is the most commonly used cosmogenic nuclide. ²⁶Al is also produced 521 in quartz and is commonly processed in the same samples as ¹⁰Be. Both nu-522 clides are produced in quartz through only two mechanisms: spallation and 523 muogenic production. Muogenic production of ¹⁰Be and ²⁶Al accounts for 524 only $\sim 1.5\%$ and $\sim 2\%$ of total surface SLHL production, respectively, as cal-525 culated using CRONUScalc. CRONUScalc employs the currently accepted 526 half-life and AMS standard values for ¹⁰Be (Nishiizumi et al., 2007; Ko-527 rschinek et al., 2010; Chmeleff et al., 2010). The online interface performs 528 automatic renormalization for all ¹⁰Be and ²⁶Al concentrations based on the 529 standards selected by the user. The interface passes this information to the 530 underlying raw CRONUScalc program, which assumes the concentration is 531 normalized to 07KNSTD for ¹⁰Be and KNSTD for ²⁶Al. For discussions con-532 cerning these normalizations, see Balco et al. (2008) and Nishiizumi et al. 533 (2007).534

When ²⁶Al and ¹⁰Be are both being analyzed for a particular sample, they can be processed in the same quartz split. The code operates in a fashion similar to that of Balco et al. (2008) in that it allows any given sample to be associated with data from either ¹⁰Be or ²⁶Al, or both nuclides. The production rates for both nuclides have only been determined reliably in quartz, so this is the only mineral target supported by CRONUScalc at this time.

Additional information, such as erosion rate or burial history, can be determined if more than one nuclide is analyzed in a sample (Bierman, 1994; Granger & Muzikar, 2001). The common Al/Be pair is frequently used for these investigations. CRONUScalc does not currently provide figures for the paired interpretation if ¹⁰Be and ²⁶Al, but these capabilities could be added by an advanced user following the method described in Granger & Muzikar (2001); Granger (2006).

For information on the production rates used in the calculator, see the CRONUS-Earth spallation production rate calibration paper (Borchers et al., 2015) or the summary paper (Phillips et al., 2015).

552 3.2. Chlorine-36

⁵⁵³ Cosmogenic ³⁶Cl is produced by a wide range of pathways that have been ⁵⁵⁴ discussed in the production systematics section. The main three pathways are ⁵⁵⁵ spallation (Ca, K, Ti, Fe), low-energy neutron absorption (Cl), and muogenic ⁵⁵⁶ production (Ca, K, Ti, Fe). Note that we include the production of ³⁶Cl ⁵⁵⁷ by relatively low-energy neutrons (down to approximately 3 MeV) by the ⁵⁵⁸ reaction ${}^{39}K(n,\alpha){}^{36}Cl$ in the "spallation" category, using the cross-section of ⁵⁵⁹ Reedy (2013), since this energy is still well above the epithermal level.

Previous studies have resulted in production-rate estimates that differ 560 from each other by up to 40%. One goal of the CRONUS-Earth project 561 was to resolve these uncertainties (Marrero, 2012; Marrero et al., 2015, this 562 volume, submitted). For spallation production rates, only CRONUS-Earth 563 mineral separate data is used in the calibration and the resulting production 564 rates fit the independent secondary data set very well (see evaluation of the 565 secondary ³⁶Cl data set in Borchers et al. (2015)). For the low-energy neu-566 tron production pathway, however, there are additional unknown complicat-567 ing factors, potentially water content or other site-specific parameters, which 568 influenced the $P_f(0)$ calibration. A more complete discussion of calibration 569 results and factors affecting ³⁶Cl production can be found in Marrero et al. 570 (2015), but the resulting value for $P_f(0)$ fits the secondary data set well and 571 is used in CRONUScalc until additional ³⁶Cl calibrations can be performed. 572 The $P_f(0)$ value that was obtained during the CRONUS-Earth calibration 573 has a larger uncertainty (about 20%) than the other ³⁶Cl production param-574 eters. Ideally, ³⁶Cl should be measured on low-chlorine mineral separates to 575 eliminate the uncertainty associated with this pathway (cf. Schimmelpfennig 576 et al. 2009). Analysis of low-chlorine mineral separates should help to keep 577 ³⁶Cl ages to a similar level of uncertainty as other nuclides. 578

It is important to note that the predicted nuclide concentrations are more 579 sensitive to certain parameters than others. For example, hydrogen is the 580 most effective moderator of neutron energy (Fabryka-Martin, 1988), and thus 581 the amount of water in a sample can strongly affect the magnitude of the 582 low-energy neutron flux and consequently influence the production of ³⁶Cl 583 through the low-energy pathway. The importance of water was evaluated 584 theoretically by Dep et al. (1994) and experimentally substantiated by Dunai 585 et al. (2014), indicating that the presence of hydrogen can significantly influ-586 ence production. There is some evidence to indicate that the water content 587 of the surrounding landscape may also affect the low-energy neutron flux 588 (Zreda et al., 2008). The sensitivity of sample age to water in the current 580 calculator is included in the sensitivity study in Section 5.2. 590

⁵⁹¹ Chlorine-36 dating requires compositional parameters that most cosmo-⁵⁹² genic nuclides do not. The code allows for the input of both "mineral target" ⁵⁹³ and "bulk-rock" compositions. The bulk-rock analysis permits the calcula-⁵⁹⁴ tion of the neutron-transport parameters. If a mineral separate has been used

for the analysis, the target-element concentrations (comprehensively, K, Ca, 595 Ti, Fe, and Cl) are also needed for the sample on which the measurement 596 was performed. The bulk-rock composition includes data on trace-elements, 597 such as boron and uranium, that are needed for the appropriate calculation 598 of neutron-absorption properties of the rock and radiogenic subtraction. For 599 samples with low Cl content and high concentrations of K and/or Ca, the 600 full range of trace element analyses may be superfluous. Given a complete 601 analysis of a rock sample, CRONUScalc calculates the percentage of the pro-602 duction originating from low-energy neutron absorption by Cl, spallation on 603 Ca and K, and muons. This can be used to determine whether chemical 604 analysis of elements that modulate the low-energy neutron flux are needed 605 for every sample. 606

Isotope Dilution Mass Spectrometry (IDMS) is a standard method for 607 highly accurate elemental and isotopic analysis. In cosmogenic-nuclide ap-608 plications it is commonly used for analysis of ³⁶Cl and elemental Cl. The 609 process includes adding a "spike" of known isotope ratio that has been en-610 riched in one of the stable chlorine isotopes, either ³⁵Cl or ³⁷Cl. In either case. 611 the ratios of ³⁶Cl/³⁵Cl and ³⁵Cl/³⁷Cl are measured by the accelerator mass 612 spectrometer. The stable chloride concentration and the ³⁶Cl concentration 613 of the rock can both be back-calculated using the ratios from the accelerator 614 and the recorded spike and sample masses from sample preparation. The 615 process is described in detail in Desilets et al. (2006a). 616

When using IDMS for chlorine, there is a correlation between the uncer-617 tainties in stable chloride concentration and the uncertainties on 36 Cl con-618 centration because they both depend on the same isotope ratios. In order to 619 produce the correct uncertainties from raw laboratory results, a set of addi-620 tional codes were created to handle this calculation. This code is external 621 to the main CRONUScalc program, but is distributed with the code. It is 622 designed to assist the user in calculating the correct concentrations of ³⁶Cl 623 and Cl and correctly assigning their uncertainties for samples reported as 624 ³⁶Cl/total Cl and ³⁵Cl/³⁷Cl by AMS. A linearized uncertainty method was 625 used to calculate the stable chlorine concentration (spiketoconc.m). How-626 ever, the traditional error propagation does not work for low-Cl samples due 627 to the non-linear response in this range, so uncertainties are calculated using 628 a Monte Carlo method (spiketoconcmc.m). In the Monte Carlo code, the 629 uncertainty in the stable chlorine concentration is calculated 10,000 times us-630 ing the nominal inputs (spike mass, sample mass, spike concentration, etc.) 631 and a random uncertainty. The random quantity added to the nominal value 632

of the parameter is normally distributed with a mean of 0 and standard deviation equal to the uncertainty in the parameter. For samples with high chlorine concentrations, the Monte Carlo result is essentially the same as the linearized approximations; however, at low chlorine concentrations, the uncertainties can be a significant percentage of the total concentration (>90%).

638 3.3. Carbon-14

Production of in-situ cosmogenic ¹⁴C is primarily from the spallation of 639 oxygen, but also other elements including Mg, Al, and Si (Dunai, 2010). 640 Unlike 10 Be, muons contribute significantly (15-20%) to the cosmogenic pro-641 duction at the surface and increasingly at depth. Quartz is the only mineral 642 phase in which ¹⁴C is commonly measured, although it is possible to measure 643 it in other minerals as well (Handwerger et al., 1999; Dunai, 2010). Carbon-644 14 has a short half-life and therefore reaches secular equilibrium between 645 production and decay relatively quickly ($\sim 25-30$ kyr) (Lifton et al., 2001). 646 Due to these factors, ¹⁴C is well-suited for short-term erosion rate studies 647 using multiple nuclides and young burial-history studies (Dunai, 2010). 648

In situ ¹⁴C is extracted from quartz typically by stepped combustion/ dis-649 solution (e.g. Lifton et al., 2001; Pigati et al., 2010; Goehring et al., 2014) or 650 stepped combustion (Hippe et al., 2014). Reproducibility of intercomparison 651 materials is typically ca. 5% among labs (Jull et al., 2013), but occasional 652 discrepancies on replicate analyses have been as much as 5-10% in the past 653 (Borchers et al. 2015). As a result, in situ ¹⁴C production rates have tended 654 to exhibit somewhat greater uncertainties than other nuclides, although the 655 situation is improving as extraction techniques continue to advance. 656

In CRONUScalc, only the production of ¹⁴C in quartz by spallation and by muons is available. This should provide adequate functionality for the majority of ¹⁴C users. For information on the ¹⁴C production rates used in the calculator, see the CRONUS-Earth spallation production rate calibration paper (Borchers et al., 2015).

662 3.4. Helium-3

Cosmogenic ³He is produced primarily by spallation reactions on most elements present in a rock (Mg, Si, Fe), but also through low-energy neutron absorption by lithium (Li) (Dunai et al., 2007). Moderate amounts of Li (50-200 ppm) can cause a significant amount of production to originate from cosmogenic thermal neutrons, affecting neighboring (and potentially Li-free) mineral phases (Dunai et al., 2007). Even in relatively low-Li samples (1

ppm), radiogenic production can be important in rocks where the formation 669 age of the rock is much greater than the exposure age of the sample (Lal, 670 1987; Farley et al., 2006; Dunai et al., 2007). There is currently no evidence 671 of significant contribution to production via muon reactions (Kurz, 1986: 672 Dunai, 2010). The most commonly used minerals are pyroxene and olivine, 673 however ³He has been measured in other mineral phases (e.g. apatite, zircon, 674 biotite, titanite, kyanite, and hornblende) (Amidon & Farley, 2012; Amidon 675 et al., 2009; Amidon & Farley, 2010; Farley et al., 2006; Amidon et al., 2008). 676 When evaluating ³He data, the inherited (magmatic) component must 677 be subtracted from the measured concentration in order to include only the 678 cosmogenically produced ³He in the dating. This is done through methods 679 that vary by lab (Kurz, 1986; Cerling, 1990; Blard & Pik, 2008; Blard & 680 Farley, 2008; Williams et al., 2005). The input needed for CRONUScalc is the 681 cosmogenic concentration of ³He in units of atoms per gram. CRONUScalc 682 assumes that any corrections for blanks or inherited component have already 683 been performed. 684

Currently, CRONUScalc only incorporates spallation production for ³He. 685 The spallation production rate is currently calibrated from a combined data 686 set consisting of olivine and pyroxene samples from the compilation by Goehring 687 et al. (2010b). The publication includes both official CRONUS data and 688 high-quality external data sets. Individual sites were determined to be either 689 primary or secondary data sets based on quality of the independent age con-690 trol and other site parameters. For information on the ³He calibration, see 691 Borchers et al. (2015). 692

Recommended use of the calculator is currently limited to olivine and 693 pyroxene due to the limitations of the calibration data set. For pyroxene 694 and olivine derived from basalt and xenoliths, Li concentrations are typically 695 very low (<5 ppm) so the presence of cosmogenic or radiogenic ³He from 696 low-energy neutrons should be negligible (Dunai et al., 2007). The lithium 697 low-energy pathway needs to be considered for minerals in rocks that are 698 more evolved than primitive basalts and generally for rocks whose geological 699 age is much larger than the exposure age (Dunai et al., 2007; Lal, 1987). 700 Until this pathway is incorporated into the program, users are cautioned to 701 limit the use to low-Li samples. 702

The calculation of the low-energy production pathway is complicated by the large number of additional parameters that would be needed (e.g. grain size of target minerals, complete major and trace element composition of the host rock, water content) (Dunai et al., 2007). The calculator is already designed to take the mineral phase as an input and a thermal neutron component could be added by adapting the low-energy production calculations
already in place for ³⁶Cl. This would increase the applicability of CRONUScalc to helium samples with higher Li contents and ultimately allow inclusion
of other mineralogies.

712 4. CRONUScalc Calculators

The program written to perform a range of cosmogenic nuclide calcu-713 lations is named "CRONUScalc." It is written in the MATLAB program-714 ming language, but can also be run under open-source Octave. The pro-715 gram is available under the GNU General Public License agreement and can 716 be accessed and downloaded at: https://bitbucket.org/cronusearth/ 717 cronus-calc. In the repository, users can upload their own files alongside 718 the main code in order to share new features or functions using CRONUScalc. 719 The program is organized by scaling model, with folders containing code 720 to perform various calculations. The core functions (Section 4.1) and the 721 functions common to all nuclides are contained in the **production** folder. Ad-722 ditional folders are available for surface sample dating (surfacecalc), depth 723 profile dating (profilecalc), and calibration of production rates (calib, 724 muoncalib, pfzero). 725

726 4.1. Core Functionality

The code is designed to predict concentration at a particular depth for 727 the specified nuclide. This is done using a function called **predNXX**, where 728 XX represents the desired nuclide (i.e. predN36 calculates production of 720 ³⁶Cl). There are a few preliminary code modules necessary to build the in-730 puts required for the main code. These include creating constants for physi-731 cal parameters (physpars), calculating scaling factors (scalefacsXX), orga-732 nizing the input sample parameters (sampparsXX), and computing sample-733 dependent parameters that are needed for later calculations (compparsXX). 734 These are not discussed in detail here, but can be found in the Function 735 Appendix in the Supplementary Materials and will be available online for 736 download with the code. PredNXX uses all this preliminary information in 737 order to predict concentrations of the nuclide. This is summarized in Figure 738 3 for the 10 Be code. 739

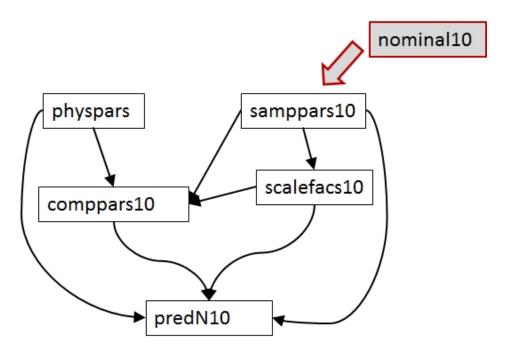


Figure 3: Data flow diagram showing the relationship between different parts of the code for a ¹⁰Be sample. Inputs include the nominal inputs (nominal10) and the uncertainties on each of those inputs (uncerts10, not shown). Other code pieces (physpars, samppars10, comppars10, and scalefacs10) set up the correct variables for the upper-level code pieces. predN10 predicts the concentration for a given age and depth.

740 *4.2.* Features

This program has several differences as compared to previous calculators. 741 The CRONUScalc program is designed to work for all the commonly used 742 nuclides, including ¹⁰Be, ²⁶Al, ³He, ³⁶Cl, and ¹⁴C, with ²¹Ne in testing. The 743 modular nature of the program means that the scaling frameworks are ap-744 plied in an identical manner to each nuclide, avoiding possible inconsistencies 745 from errors in coding or using two different calculators for cross-nuclide com-746 parisons. The addition of a new scaling framework by Lifton et al. (2014)747 allows for a physics-based calculation of the incoming cosmic-ray flux as well 748 as nuclide-dependent scaling frameworks that incorporate the individual nu-749 clide reaction cross-sections. This calculator has an easy input structure so 750 that samples can be copied from spreadsheets and allows the user to run 751 multiple samples simultaneously. This is one of the only calculators to allow 752 the user to specify uncertainties on all input parameters and propagate those 753 to the resulting exposure age. Finally, this program does not require any pro-754 prietary software because it can be run on either MATLAB or Octave, the 755 latter being open-source. 756

CRONUScalc is able to calculate production and accumulation for any given sample as well as perform calibrations and calculate surface sample and depth-profile exposure ages. This calculator provides both surface sample and depth-profile dating abilities based on the same underlying code. Included in the surface calculator is the ability to date a single sample at depth. The code does not currently calculate burial ages.

Other new features that are not clearly visible in the upper levels of the 763 program are the new integration method and the newly calibrated Heisinger 764 muon production formulation. Some previous calculators relied on analytical 765 solutions to integrate over depth by incorporating a 'thickness correction 766 factor'. In this program, a numerical depth integration is performed, which 767 removes approximations necessary in the analytical solution. This results in 768 a more accurate inventory of accumulated nuclide in the sample as well as 769 the ability to easily modify the code to look at multiple time periods with 770 varying erosion rates. The new muon production is scaled using Lifton et al. 771 (2014), and includes new production rate parameters calibrated from deep 772 profiles for ¹⁰Be, ²⁶Al, and ³⁶Cl (for both Ca and K). 773

Finally, although this paper discusses primarily the code behind the program, there is an online interface for the surface calculator, and other small tools such as the topographic shielding calculator, available to the public with no need to directly manipulate the code (http://web1.ittc.ku.edu: 8888/). This simplifies the process and makes the technique available to a
much broader range of users. Instructions for use of the interface calculators
will be maintained online.

781 5. Surface Sample Calculator

The Balco et al. (2008) calculator put the Al/Be cosmogenic nuclide community on a consistent platform by providing a reliable tool for consistent ¹⁰Be and ²⁶Al exposure age calculations. However, this did not apply to any other nuclides and the code did not allow the user to fully propagate uncertainties on all input parameters. CRONUScalc extends this idea to include multi-nuclide capability, full propagation of uncertainties, and the ability to work with single samples at depth.

Using the equations given in the previous sections, we can compute the 789 rate of production of a cosmogenic nuclide at any depth within the subsur-790 face at any point in the past. This production rate varies over time due to 791 time-dependent scaling as well as varying due to changes in depth caused 792 by erosion or aggradation. While the nuclide is accumulating it is also con-793 tinuing to decay - the radioactive decay rate must be subtracted from the 794 production rate. The time-dependent production rate can be numerically 795 integrated over time in order to predict the concentration of the cosmogenic 796 nuclide at a specific depth after a specified exposure history. By averaging 797 the accumulated production at depths throughout the thickness of a sam-798 ple, we can predict the average concentration of the cosmogenic nuclide as a 790 function of the exposure age of the sample. 800

⁸⁰¹ 5.1. Computing the Exposure Age or Erosion Rate

To compute the exposure age of a sample, we begin by checking for sat-802 uration. This is done by computing the sample-specific saturation concen-803 tration by predicting the accumulated concentration in the sample assuming 804 an exposure time of at least six half-lives, incorporating time-dependent pro-805 duction rates and ending in the sample collection year, and using all other 806 user-provided inputs (see Section 3 for a list of inputs). If the measured 807 concentration of the cosmogenic nuclide is close to or exceeds the satura-808 tion concentration (95% or more), then it is not possible to determine an 809 exposure age for the sample because the sample is effectively saturated. If 810 not, then we use the bisection method to find an age at which the predicted 811 concentration matches the measured concentration of the nuclide. A similar 812

method is applied in order to determine erosion rate for a sample when either an independent age is supplied or equilibrium with erosion rate can be assumed.

⁸¹⁶ 5.2. Input Uncertainties and Derivatives

The computed exposure age for a sample depends on a number of sample 817 parameters in addition to the measured nuclide concentration, including its 818 thickness and density, the assumed erosion rate, the assumed atmospheric 819 pressure at the exposure site, etc. For each of these parameters, we compute 820 the derivative of the exposure age with respect to the parameter by finite 821 difference approximation. If the user supplies uncertainties for these param-822 eters, then these derivatives are employed to propagate the uncertainties in 823 the parameters into an uncertainty on the computed exposure age, using first 824 order Taylor series expansion (Bevington & Robinson, 1992). 825

The code allows for detailed quantification of the uncertainties associ-826 ated with a sample. The commonly reported uncertainty, that from only the 827 AMS analysis, represents the minimum uncertainty in the age of an unknown 828 sample. There are uncertainties in the other measurements (chemical concen-829 trations, field measurements, etc.), production rates, scaling, and laboratory 830 processing. CRONUScale allows an uncertainty to be assigned to each of the 831 input parameters, a feature that is unavailable in other single-sample sur-832 face exposure age calculators. This idea was addressed in the multi-sample 833 depth-profile calculator using Monte Carlo methods presented by Hidy et al. 834 (2010), but is treated more systematically here using derivatives. In a manner 835 similar to that employed by Balco et al. (2008), CRONUScalc distinguishes 836 between analytical (internal) uncertainty, which is calculated by propagating 837 uncertainties on every input parameter in uncertsXX, and total uncertainty, 838 which is calculated by fully propagating the production-rate uncertainty and 839 combining it with the analytical uncertainty. Although statistically-based 840 uncertainties on the production rate parameters were not possible (Borchers 841 et al., 2015), an alternative method to assess the uncertainty on this parame-842 ter was used in an attempt to provide a more complete uncertainty estimate. 843 The uncertainty associated with the nuclide production rate (i.e., the 844

uncertainty added to the analytical uncertainty to give the total uncertainty) is based on a comparison of the deviations of the calculated ages for the secondary data set with the independent (and assumed correct) ages for those samples. In essence, the uncertainty associated with the production rate was increased until the average standard deviation of the calculated ages matched

the spread of the deviations of the calculated ages from the independent 850 ages. This empirical estimation of the additional uncertainty assumes that 851 the residuals between the calculated ages and the independent ages averages 852 zero and has a normal distribution. The actual distribution of residuals only 853 marginally supports this assumption. The most significant sources of the 854 additional uncertainty (not accounted for by the assigned analytical or other 855 measurement uncertainties) are the following four. (1) Underestimation of 856 the actual analytical uncertainties. Jull et al. (2013) have presented evidence 857 that laboratory-reported uncertainties generally underestimate, sometimes 858 by significant amounts, the actual spread of the cosmogenic-nuclide analytical 859 data. (2) Errors in spatial scaling. Regularities in the residuals as a function 860 of location and elevation give clues that even the best scaling models may 861 not completely predict the global pattern of nuclide production (Phillips 862 et al., 2015). (3) Errors in the assigned independent ages for the primary 863 calibration data sites. Although sites with very well constrained ages were 864 used, they cannot be guaranteed to all be completely accurate. (4) Site-865 specific factors. Assumptions regarding factors such as snow cover, lack of 866 covering deposits in the past, and erosion rates may have been in error for 867 some sites. In summary, the additional uncertainty that is specified for the 868 production-rate terms incorporates all sources of uncertainty or bias that are 869 not included in the reported laboratory analytical uncertainties. It reflects 870 the likelihood that a single, randomly selected, high-quality, cosmogenic-871 nuclide age will correspond to an independently determined exposure age for 872 the same material, empirically assessed using the CRONUS-Earth secondary 873 data sets. Full details on the method, assumptions, and the uncertainty 874 sources incorporated into the production-rate uncertainty are discussed in 875 Phillips et al. (2015). 876

It is important to differentiate between the two reported uncertainty types 877 (analytical and total). For example, analytical uncertainties alone are likely 878 to underestimate realistic uncertainties when comparing to other dating tech-879 niques or comparing widely geographically or temporally separated cosmo-880 genic samples. On the other hand, the total uncertainty may overestimate 881 uncertainties when comparing between groups of samples from a single ge-882 ographic location where uncertainties are not independent (e.g. in certain 883 cases, production rate uncertainties are unimportant for relative chronology 884 at a single site) (Dunai, 2010; Balco et al., 2008). Comparisons between nu-885 clides can be complicated by the use of different calibration data sets and 886 production models, so it may be necessary to use the external uncertainties 887

to compare between samples from different nuclides located at the same site (Balco et al., 2008). Both uncertainties are reported, allowing the user to determine which is the most appropriate for each particular study. The user is encouraged to report all inputs, including assigned uncertainties, and note the type of uncertainty used in analyses.

A sensitivity analysis was performed in order to determine which inputs 893 and their uncertainties have a significant effect on the sample exposure age. 894 The results of the sensitivity analysis are presented in Figure 4. It is based 895 on a random selection of the ³⁶Cl primary and secondary calibration samples 896 (see Marrero (2012) for details), although similar results were seen for ^{10}Be 897 samples. The results presented are merely intended to be representative and 898 will vary from sample to sample. Sensitivities are only given for standard 899 environmental parameters that are applicable for all nuclides. Chlorine-36 900 uncertainties are also sensitive to sample chemical composition. This topic 901 is dealt with separately for 36 Cl in Marrero et al. (2015). 902

Based on this sensitivity analysis, there are some uncertainties that are in-903 significant. For example, realistic uncertainties on longitude and latitude are 904 not important when compared to uncertainties on the concentration. Other 905 variables that realistically do not require uncertainties include shielding and 906 year collected. On the other hand, relatively large and potentially biased 907 uncertainties are expected on parameters such as erosion rate and water con-908 tent because these require estimation and some knowledge of the site and 909 can rarely be precisely calculated. 910

When comparing different nuclide results, especially those from different 911 sites, all uncertainties must be assessed in a consistent manner. The un-912 certainties from scaling and methodological considerations become primary 913 issues. The interlaboratory studies associated with the CRONUS project 914 (Jull et al., 2011, 2013; Vermeesch et al., 2012; Schaefer et al., 2014; Blard 915 et al., 2014) were designed to address the uncertainties that arise solely from 916 different processing techniques and accelerator measurements. These results 917 were used to incorporate realistic uncertainty into the nuclide concentrations 918 used for calibration (see Phillips et al. 2015 for details). 919

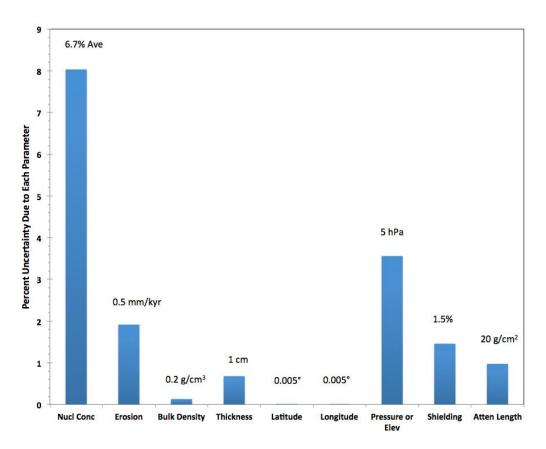


Figure 4: Amount of uncertainty (as a percentage of the final calculated age) contributed by conservative uncertainties assigned to individual sample parameters. The analysis presents the average of the propagated uncertainties for 15 primary and secondary ³⁶Cl calibration samples (Marrero, 2012; Marrero et al., 2015). The values above each bar give the assigned uncertainty in the parameter.

920 6. Depth-Profile Calculator

The depth profile calculation is formulated as a Bayesian inverse prob-921 lem. This approach has several advantages. First, unlike using a classical 922 statistics approach to parameter estimation, the Bayesian approach treats un-923 known parameters as random variables. Doing so allows the resulting fitted 924 parameters to be reported as expected values, accompanied by a probability 925 distribution. Second, this approach allows the user to submit prior infor-926 mation or expert knowledge, influencing the solution. The selection of an 927 informative prior distribution for parameters will strongly bias the resulting 928 posterior distribution. Therefore the responsible user is obligated to report 929 the effect that different priors have on their solution. Third, from a compu-930 tational standpoint, the method presented in this section is not susceptible 931 to the convergence failures that iterative line-search solvers can encounter in 932 certain cases. 933

The inputs to the depth profile code are the same as those for the surface 934 calculator, with several additional parameters required only for the profile 935 calculation. The new parameters include maximum and minimum values 936 for erosion rate, exposure age, and inheritance. In addition to erosion rate 937 bounds, a value for total maximum erosion (positive or negative) can be used 938 to additionally constrain the final results, which can be useful as total erosion 939 can sometimes be easier to determine in the field. Some parameters common 940 to the surface calculator, such as depth to top of sample and attenuation 941 length, become more significant in the depth profiles than they were for 942 surface samples. These parameters are both user inputs (i.e. the calculator 943 assumes these are known parameters), although the uncertainties in these 944 parameters have the potential to increase the uncertainty in the calculated 945 age of the profile. One initial sensitivity test on a profile indicated that 946 reasonable uncertainties on attenuation length were relatively insignificant, 947 although reasonable uncertainties on bulk density measurements could result 948 in up to 5% change in nominal age. For details on other parameters, see the 949 sensitivity study results in Section 5.2. The depth profile calculator does not 950 explicitly incorporate uncertainties on input parameters other than nuclide 951 concentration. 952

Performing the depth profile calculation requires the simultaneous estimation of three parameters: age (t), erosion rate (ϵ), and inheritance (inh). The steps necessary for the computations are summarized here. First, an evenly spaced, 3-dimensional grid over the parameters of age, erosion rate, and inheritance is created. Note that the spacing is consistent only within each dimension and the parameter range for each is specified by the user. Second, the misfit χ^2 value is calculated at each node (each age, erosion rate, and inheritance point) using Equation 10.

$$\chi^2_{t,\epsilon,inh} = \sum_{i}^{n} \left(\frac{x_i}{\sigma_i}\right)^2 \tag{10}$$

where

$$x_i = ConcPred_{t,\epsilon,inh} - ConcMeas_i$$

Next, the approximated χ^2 hyper-surface is transformed into a likelihood surface using Equation 11.

$$L(\mathbf{x}|\boldsymbol{\theta}) = \prod_{i=1}^{n} \left(\frac{1}{\sqrt{2\pi\sigma_i}}\right) \cdot \exp\left(-\frac{\chi_{t,\epsilon,inh}^2}{2}\right).$$
(11)

Finally, to calculate the joint posterior from the likelihood we use Bayes rule, shown in Equation 12.

$$p(\boldsymbol{\theta}|\mathbf{x}) = \frac{L(\mathbf{x}|\boldsymbol{\theta})\pi(\boldsymbol{\theta})}{\int_{-\infty}^{\infty} L(\mathbf{x}|\boldsymbol{\theta})\pi(\boldsymbol{\theta})d\boldsymbol{\theta}},$$
(12)

where $\pi(\boldsymbol{\theta})$ is the joint prior distribution set by the user on the following parameters: age, erosion rate, and inheritance. A trapezoidal integration scheme is used to calculate the denominator, leaving the joint posterior density, $p(\boldsymbol{\theta}|\mathbf{x})$.

When assigning uncertainties to a solution, it is important to determine 969 if any multiple interactions exist between erosion rate, age, and inheritance. 970 To display these interactions, the calculator produces 3 pairwise 2-D contour 971 plots of the joint posterior distribution with contours of 68% and 95% regions 972 of probability. For example, integrating $p(\boldsymbol{\theta}|\mathbf{x})$ over inheritance gives the 973 probability distribution given by $p(\epsilon, t | \mathbf{x})$. This effectively marginalizes the 974 joint posterior over one of the three parameters. An example of the resulting 975 2-D contour plots are shown in Figure 5, along with the predicted versus 976 measured profile plot. 977

The best-fitting solution is the lowest chi-squared value over the entire three-dimensional grid. This is called the maximum a posterior (MAP) solution. In the 2-D pairwise plots, the MAP solution and the apparent 2-D bestfit solution do not always coincide due to the interaction of the parameters

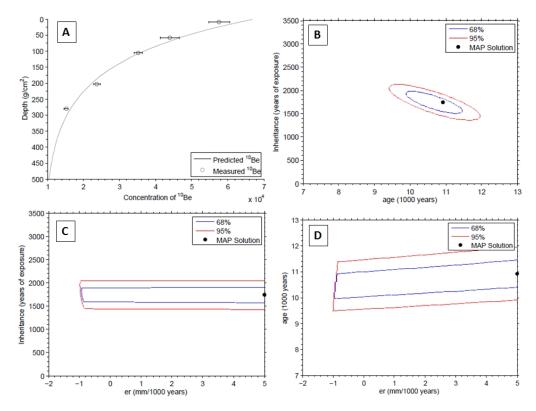


Figure 5: Example of the profile and pairwise plots produced by the depth profile calculator. The example is a ¹⁰Be profile published by Goehring et al. (2010a) and used as an example in Aumer (2010). MAP solution is the best-fitting 3-D solution. 68% and 95% contours are the confidence intervals for the chi-squared values. (A) Figure shows the measured data and the predicted profile with depth; (B) Figure shows the confidence intervals for the plot of the inheritance and age; (C) Figure shows the confidence intervals for the plot of the inheritance and erosion rate; (D) Figure shows the confidence intervals for the plot of the plot of the age and erosion rate.

with the third dimension. The direct calculation of posterior probabilities 982 eliminates the need for Monte Carlo, as was used by Hidy et al. (2010). In 983 contrast to Hidy et al. (2010), this code also computes a posterior probabil-984 ity distribution, allowing for an assessment of the probability of the resulting 985 age being within particular age bounds. While there are other mathematical 986 methods or spreadsheets available for calculating depth profiles (Braucher 987 et al., 2009; Hein et al., 2009; Schaller et al., 2009), we discuss CRONUScalc 988 in relation to Hidy et al. (2010) because of its similar format and explicit 989 distribution for use as a depth profile calculator. 990

991 7. Discussion and Cautions

992 7.1. Atmosphere and Elevation Relationships

The relevant inputs are both elevation and atmospheric pressure for the 993 sample. Elevation, which is more easily measured and can be found on a to-994 pographic map even after sampling, is the traditional input for these codes. 995 The additional pressure input is necessary because it is the more accurate 996 measure of the sample location in the atmosphere. If the user does not specify 997 a site pressure, the online user interface automatically estimates the pressure 998 using the ERA40 Reanalysis data set. The ERA40 reanalysis is a synthesis of 999 comprehensive global observations and analyses over a 45 year period, as dis-1000 cussed in Uppala et al. (2005). This pressure calculation module is available 1001 with the code and pressure is a required input (not automatically calculated) 1002 within the raw code. The production-rate calibrations were all done using 1003 pressures determined from ERA40 and it is recommended that this conver-1004 sion be used unless there is a specific reason to suspect a significantly different 1005 pressure relationship over the sample exposure period. ERA40 functions in 1006 Antarctica so no separate atmosphere relationship is required. 1007

Uncertainty in the sample pressure is a significant contributor to sample 1008 uncertainty. The uncertainty associated with the pressure based on uncer-1009 tainties in elevation is quite small (<1 hPa for elevation uncertainties up to 1010 15 m). This results in only a small amount of total uncertainty on the sample 1011 age (<1%). A more realistic source of uncertainty associated with pressure 1012 is the difference between the current pressure and the average pressure over 1013 the exposure time of the sample. The pressure history through time is not a 1014 factor that can be accurately calculated. Our assumption that the pressure 1015 has remained relatively constant through time adds additional uncertainty. 1016 Based on the maximum differences observed between corresponding grid cells 1017

in the NCEP/NCAR and ERA40 (Uppala et al., 2005) reanalyses, we have added a conservative uncertainty of 5 hPa to the CRONUS-Earth samples to account for this and this addition results in uncertainties of 2-5% for most samples.

1022 7.2. Uncertainties Due to Scaling

Not all scaling frameworks fit the calibration data equally well. For each 1023 nuclide, a single data set was used to produce a production rate for each com-1024 bination of scaling framework and nuclide. Because of its favorable analytical 1025 precision and accuracy, relative simplicity of its production mechanisms, and 1026 the large number of analyses, the nuclide best suited to evaluating the ad-1027 equacy of scaling frameworks is ¹⁰Be. This topic is discussed more fully in 1028 Borchers et al. (2015) and Phillips et al. (2015), but is briefly summarized 1029 here. The average absolute error, by site averages, of the predicted vs. mea-1030 sured concentrations for the neutron-monitor-based scaling methods (Lifton, 1031 Dunai, Desilets) varied from 15.7 to 18.1 %. Those for the scaling methods 1032 based on more direct measures of spallation reactions (Lal-based and LSD) 1033 varied from 8.5 to 9.7 %. The second group is clearly more accurate. Al-1034 though within this group the LSD-flux based model gave the best fit, the 1035 differences are so small that no clear preference is evident. In the following 1036 discussion of scaling uncertainty, the neutron-monitor based scaling methods 1037 were not considered due to the poor fit to the data set. 1038

A second issue associated with scaling is the uncertainty from determining 1039 the scaling factor at a new location that is not a calibration site. The basis 1040 for estimating this contribution to the total age uncertainty is weak. The 1041 best basis available is found by comparing the fit of the primary CRONUS-1042 Earth ¹⁰Be data set (to which the production rates were calibrated) to the 1043 fit of the secondary data set (which is independent of the primary data set) 1044 in Borchers et al. (2015). For the best-fitting scaling framework (LSD-flux 1045 based), the average absolute error was 5.4 % for the primary data set, com-1046 pared to 8.5 % for the secondary data set. The difference between these two 1047 uncertainties could be interpreted to suggest that error in predicting scaling 1048 at non-calibration sites contributes about 3 % to the total age uncertainty. 1040 However, the following factors must be considered: (1) the source of the 1050 variability cannot be assigned strictly to scaling and could, in fact, be due 1051 to other issues, such as the production rate calibration (2) samples were as-1052 signed to the secondary data set because they were weaker than those in the 1053 primary one due to factors such as less well constrained exposure histories, 1054

and therefore there are undoubtedly factors in addition to scaling uncer-1055 tainty contributing to their larger scatter, and (3) error in scaling clearly 1056 contributes to the 5 % scatter obtained for the primary data set, giving a 1057 contribution to the uncertainty from scaling that is not accounted for in the 1058 comparison. Factors (1) and (2) contribute to a decrease in scaling uncer-1059 tainty while factor (3) contributes to an increase in uncertainty. The balance 1060 between these cannot at present be known, but likely the error in scaling, on 1061 average, contributes significantly less than 3 % to the total age uncertainty. 1062 The original CRONUS-Earth production-rate calibration was not able to 1063

calculate statistical uncertainties on the production rate parameters (Borchers 1064 et al., 2015). Instead, the CRONUS-Earth Project employed an empirical 1065 method for estimating production-rate uncertainty based on the secondary 1066 data set (Phillips et al., 2015). The empirical method does not break out the 1067 contribution to that uncertainty from scaling alone, but it does incorporate 1068 that source of uncertainty into the production-rate uncertainty recommen-1069 dation. This inferred production-rate uncertainty is propagated along with 1070 other parameter uncertainties in CRONUScalc to provide the total uncer-1071 tainty on a sample. Additional insight into scaling uncertainty can be ob-1072 tained from the "leave-one-out cross-validation exercise" in Borchers et al. 1073 (2015).1074

1075 7.3. Limitations of CRONUScalc

Although CRONUScalc has new capabilities not previously available in 1076 other calculators, there are some limitations to the program. CRONUScalc 1077 was designed to provide the most accurate model for a wide range of samples, 1078 meaning that approximations were avoided in most cases unless they could 1079 be shown not to increase uncertainty or error. The incorporation of more 1080 physics-based modeling into the program has substantially increased the cal-1081 culation time for each sample. The online interface minimizes this problem 1082 by emailing the user with results, original inputs, and all data necessary 1083 to recreate the graphs prior to their deletion from the server so no further 1084 contact with the website is required. While samples are running, the user 1085 is provided a link that shows current progress on submitted samples. The 1086 use of email avoids browser timeout problems and decreases the dependence 1087 on a reliable internet connection. Emails and all sample data, except usage 1088 statistics on general location and nuclide, are automatically deleted every 24 1089 hours from the server to protect anonymity. 1090

The current program only process one scaling model at a time for each sample. While it can be instructive to examine outputs from multiple scaling models, the addition of two new scaling models, for a total of seven, makes this time-prohibitive given the new framework. By allowing multiple samples to be submitted simultaneously, users can still perform the scaling comparisons by submitting samples more than once with a different scaling model selection.

¹⁰⁹⁸ 7.4. Expected Changes in Performance Due to New Scientific Content

Comparisons to results from other similar cosmogenic calculators do not 1099 serve to show that CRONUScalc functions as expected for two main rea-1100 sons. First, differences in the calibration data sets used by each program 1101 lead to very different results. Second, CRONUScalc has a different set of as-1102 sumptions, parameters, and implementations than other programs, leading 1103 to results that differ by more than rounding error from the other programs. 1104 even when identical calibration data sets are used. Fundamentally, differences 1105 between the various programs are expected and this makes it impossible to 1106 prove the accuracy of CRONUScalc by obtaining the same results as those 1107 produced by other programs. The main differences are discussed later in this 1108 section. 1109

However, in the very simplest case where there is only spallogenic pro-1110 duction and we assume a time-independent production rate, it is possible to 1111 compare results from our code with the analytical solution given by equation 1112 4.11 in Dunai (2010). The ³He calculator was tested against analytically 1113 predicted concentrations for sample ages varying between 0.001-7000 ka and 1114 erosion rates of 0, 1, and 10 mm/kyr. At the highest erosion rates, the oldest 1115 samples were saturated. For all unsaturated samples, the differences between 1116 the analytically produced concentrations and the CRONUScalc solution were 1117 less than 0.005%, with most several orders of magnitude smaller. The results 1118 are presented in Appendix D. The agreement between CRONUScalc and 1119 analytical solutions indicates that the CRONUScalc numerical integration 1120 performs well over a variety of erosion rates and ages. CRONUScalc results 1121 for other nuclides cannot be directly compared to analytical solutions due to 1122 the complex muon model and other new features, but the underlying age in-1123 tegration will be as accurate as demonstrated for 3 He, because of the shared 1124 numerical integration code. 1125

The section below highlights the new scientific content in CRONUScalc that is most likely to contribute to differences in performance when compared to other similar calculators. This section does not discuss the features of other
calculators, but reiterates the key scientific content present in CRONUScalc
and the situations that may yield major differences.

The addition of a new pair of scaling models (flux-based and nuclide-1131 dependent models from Lifton et al. (2014)) provides a flexible, physics-1132 based model for scaling production rates, but it also introduces the possi-1133 bility for differences that vary by location and elevation when compared to 1134 results from other scaling models available in CRONUScalc or other pro-1135 grams. Lifton et al. (2014) contains a more detailed discussion of differences 1136 between resulting scaling factors, including the effects of solar modulation 1137 and the updated geomagnetic history. 1138

¹¹³⁹ CRONUScalc allows the user to specify an uncertainty for each parameter ¹¹⁴⁰ in the input. In addition to those analytical uncertainties, the production-¹¹⁴¹ rate uncertainty is propagated through to produce a total uncertainty for each ¹¹⁴² sample (see Sections 5.2 and 7.2). Although the added flexibility to include ¹¹⁴³ an uncertainty on each input is useful, especially for older samples where ¹¹⁴⁴ fewer parameters were accurately reported, the CRONUScalc uncertainties ¹¹⁴⁵ will likely be different from those reported by other available calculators.

CRONUScalc muon production follows the Heisinger model, but uses 1146 CRONUS-Earth calibrated muon production rates (Fig 1 in Phillips et al., 1147 2015) and is scaled using Lifton et al. (2014). In the fast muon formulation, 1148 CRONUScalc assumes a value for α (the parameter that defines how the 1149 cross-section scales with muon energy) equals one (see Section 2.1.3). This 1150 has a small effect on the elevation dependence on production rates when 1151 compared to models that use the value of α equal to 0.75 reported in the 1152 original paper (Heisinger et al., 2002b). 1153

As discussed in Section 3.4, CRONUScalc only includes the spallation pathway for ³He production calculations. This may be appropriate for some samples, but users are cautioned to check the composition of samples to confirm that this is true. Even moderate to low concentrations of lithium (Li) in a sample can potentially contribute significant production via thermal neutron production, which is not accounted for in this model.

CRONUScalc calculations for ³⁶Cl are more complex than for other nuclides, introducing more possible contributors to differences when comparing results from other programs. CRONUScalc includes ³⁶Cl production from both fast and slow muons. For samples with higher chlorine concentrations, an updated set of elemental parameters (Table 1) may make significant differences in calculated sample results, especially for samples high in boron.

Finally, CRONUScalc assumes that the ³⁶Cl concentration is in equilibrium 1166 with the radiogenic and nucleogenic production rates in order to calculate the 1167 radiogenic subtraction (only significant for samples high in Th or U). This 1168 is unimportant for rocks with very old formation ages, but it is less clear 1169 whether this is appropriate for rocks where the formation age is equal to the 1170 exposure age. For example, based on available data for basalts, it is unclear 1171 if radiogenic production should begin at eruption or if it is in equilibrium 1172 prior to that time. Additional research is required to distinguish between 1173 these two models. 1174

1175 8. Conclusion

The CRONUScalc program, designed to predict sample concentrations at 1176 a particular depth, is intended to be versatile and work quickly for the largest 1177 number of possible applications without sacrificing accuracy. The program's 1178 abilities to calculate surface and depth-profile exposure ages, calculate ero-1179 sion rates, and perform calibrations offer many options to the user. However, 1180 the code is available under a public license, so advanced users can modify the 1181 code to work for unusual scenarios. The new features, including a new scal-1182 ing framework that performs nuclide-dependent scaling and a more accurate 1183 integration method, provide an internally consistent option for cosmogenic 1184 nuclide modeling. While the code can be adapted to many different functions. 1185 an online calculator with a simple interface has been designed for surface and 1186 depth-profile exposure age calculation (http://web1.ittc.ku.edu:8888/) 1187 in order to remove any necessity of a Matlab license or knowledge of coding 1188 and make the code easily available to the general cosmogenic isotope user 1189 community. 1190

All the previous CRONUScalc versions will be archived in the online code repository and available for calculation of previously published results.

1193 9. Acknowledgments

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1200 Appendix A. Production Models

The sections below describe the fundamental theory behind the code. This appendix is provided to summarize the systematics of the program without referring back to numerous previous publications. See the main text for summaries of the key developments of CRONUScalc.

1205 Appendix A.1. Cosmic Rays

Cosmic rays originate primarily from Milky Way galaxy supernova, but 1206 also from the sun and other energetic phenomena (Dunai, 2010; Gaisser, 1207 1990; Gosse & Phillips, 2001; Pigati & Lifton, 2004). The main cosmogenic 1208 particles reaching the atmosphere are protons (87% of the cosmic-ray flux), 1209 with a smaller portion of the comic-ray flux composed of alpha particles 1210 (12%) and other heavier nuclei (1%) (Dunai, 2010). These incoming particles 1211 have a wide range of energies, with typical energies ranging from a few MeV 1212 to 10^{20} eV (Dunai, 2010). Over long periods (10 Ma), the integrated cosmic-1213 ray flux has been shown to be constant (to within 10%) when averaged over 1214 500 kyr timescales, with the uncertainties becoming larger (up to 30% or 1215 more) when averaged over timescales of hundreds of thousands to millions of 1216 years (Wieler et al., 2013). 1217

As incoming cosmic-ray particles reach the top of the atmosphere, they interact with the earth's magnetic field. Only particles with sufficiently high energy and the correct trajectory will actually reach the earth's surface. This control on the magnitude of the cosmic-ray flux is quantified by means of the cutoff rigidity. The rigidity of a particle (R) is given by Equation A.1. In the equation for rigidity, p is the particle momentum, c is the speed of light, and e is the particle charge.

$$R = pc/e \tag{A.1}$$

The vertical cutoff rigidity, the minimum threshold for the particle energy 1225 necessary in order to penetrate the field on a path normal to the magnetic 1226 field, is the standard parameter used to organize the effect of the dipole 1227 field on the cosmic-ray flux as a function of location on the surface of the 1228 earth (Dunai, 2010; Gosse & Phillips, 2001; Lifton et al., 2005). The cutoff 1229 rigidities are related to geomagnetic latitude, having very low values near the 1230 poles $(>58^{\circ} \text{ at sea level})$, resulting in admittance of essentially all cosmic-ray 1231 particles, and increasing towards the equator (Lifton et al., 2005). 1232

The complexity of the paths of cosmic-ray particles due to interaction 1233 of looping trajectories with the solid earth results in an area where there 1234 are both allowed and forbidden trajectories in an alternating pattern known 1235 as the penumbra (Hillas, 1972). In order to determine where this region is 1236 and its effects on the cosmic-ray flux, reverse particle tracking (or trajectory 1237 tracing) can be performed for particles within 20 km of the earth's surface to 1238 determine the effective vertical cutoff rigidity of a location (Shea & Smart, 1239 1983; Dunai, 2010; Lifton et al., 2005). The effective vertical cutoff rigidity 1240 is used in scaling frameworks to parameterize the cosmic-ray flux of a site. 1241 Additional detail is given in the scaling section, Appendix C. 1242

After the primary cosmic-ray particles reach the top of the atmosphere, 1243 they interact with atmospheric particles and create a cascade of secondary 1244 particles. As these secondary particles are produced and the cascade prop-1245 agates through the atmosphere, several trends are apparent: the energy of 1246 the secondary particles decreases, the total flux of particles decreases, and 1247 the flux becomes dominated by neutrons due to the ionization losses of pro-1248 tons (Dunai, 2010). During these reactions, muons are also produced due to 1249 secondary reactions by energetic incoming particles high in the atmosphere 1250 (Eidelman, 2004). Unlike the hadronic component of the flux, the muonic 1251 flux increases and then reaches a plateau as the cascade moves down through 1252 the atmosphere because muons do not interact as strongly as neutrons and 1253 are lost very slowly through ionization (Gaisser, 1990). At the earth's sur-1254 face, muons comprise the majority of the particles in the incoming cosmic-ray 1255 flux (Lal, 1988), but contribute less to surface production than do neutrons 1256 due to their lower propensity for nuclear interactions. 1257

The energy spectrum of the neutrons that reach the earth's surface de-1258 termines the rate of cosmogenic-nuclide-producing reactions. The neutrons, 1259 which compose 98% of the nucleonic flux at the earth's surface (Dunai, 2010), 1260 have peaks in the energy spectrum at 100 MeV, 1-10 MeV, and <1 eV (Dunai, 1261 2010; Goldhagen et al., 2002). The neutron energies discussed in this paper 1262 will be categorized as high (>10 MeV), fast (0.1 to 10 MeV), epithermal 1263 (0.5 eV to 0.1 MeV), and thermal (<0.5 eV). While these conventions follow 1264 other papers in the field (Gosse & Phillips, 2001; Schimmelpfennig et al., 1265 2008), there are no consistent classifications and the actual energies associ-1266 ated with the categories may vary in other publications (Dunai, 2010). 1267

1268 Appendix B. Production Equations

The production equations for CRONUScalc are provided here. The details below are discussed in general terms for production of any nuclide, designated 'k', while details specific to a particular nuclide are discussed in later sections.

1273 Appendix B.1. Spallation

Spallation refers to the emission of a large number of nucleons (relative 1274 to the original mass of the nucleus) when an atomic nucleus interacts closely 1275 with a high-energy particle. In some cases (e.g. 3 He) a cosmogenic nuclide of 1276 interest may be one of the ejected particles. More commonly, (e.g. ²⁶Al), it 1277 is the remainder of the target nucleus. Cosmogenic-nuclide production from 1278 spallation follows a well-established exponential decrease with depth. At the 1279 surface, spallation is typically the dominant production mechanism. All the 1280 nuclides discussed in this paper are produced through at least one spallation 1281 pathway. The formula for the instantaneous production rate from spallation 1282 $(P_{s,m})$ is (Gosse & Phillips, 2001; Schimmelpfennig et al., 2008): 1283

$$P_{\rm s,m}(Z) = S_T \sum S_{el,s} P_{\rm m,k}(0) C_{\rm k} \exp\left(-\frac{Z}{\Lambda_{\rm f,e}}\right),\tag{B.1}$$

where $P_{m,k}$ is the sea-level, high-latitude production rate of species m 1284 by spallation of element k (atoms $g^{-1} a^{-1}$; S_T is the topographic shielding 1285 factor (unitless); $S_{el.s}$ is the geographical scaling factor for spallation reactions 1286 for the particular reaction of interest, including temporal variation in the 1287 production rate due to fluctuations in the geomagnetic field or solar magnetic 1288 field (unitless); C_k is the concentration of the element k (atoms $g^{-1} a^{-1}$); 1289 and $\Lambda_{\rm f,e}$ is the effective attenuation length for fast neutrons (g cm⁻²). The 1290 production is summed for all target elements k that produce nuclide m to 1291 give the total spallation production rate. 1292

1293 Appendix B.1.1. Attenuation Length

The particle attenuation length is the passage length through a medium required to attenuate the original intensity of a collimated beam of particles by a factor of e^{-1} (Gosse & Phillips, 2001). This value varies depending on the type of particle (proton, neutron, muon) and the associated energy level (fast, thermal, epithermal, etc.). For neutrons, the main factor is the energy level, with higher energy particles penetrating further into the subsurface

than lower energy particles. The apparent attenuation length, $\lambda_{f,e}$, is the 1300 flux-weighted integral of the particle attenuation length over the entire sky. 1301 The apparent attenuation length is an important parameter because it 1302 quantifies the depth distribution of the production by neutron spallation. 1303 The apparent attenuation length is defined with respect to a flat sample with 1304 no topographic shielding. When the apparent attenuation length is adjusted 1305 to account for the dip of the sample surface and any topographic shielding, 1306 the result is the effective attenuation length, $\Lambda_{f,e}$. The effective attenuation 1307 length is the parameter that should be used in calculations pertaining to 1308 production from a particular sample. 1309

Typically, as the topographic shielding increases, the effective attenua-1310 tion length increases (Dunne et al., 1999). The spectrum of neutron energy 1311 varies depending on the azimuth angle. The highest energy cosmic rays are 1312 vertically incident upon the sample and the flux is dominated by these rays 1313 (Dorman et al., 1999, in Dunai 2010). As the incident angle decreases going 1314 from vertical to horizontal, the intensity of the cosmic rays decreases due to 1315 the longer transport paths through the atmosphere (Dunai, 2010). The equa-1316 tion for this change in intensity is shown in Equation B.2. As topography 1317 typically blocks out only cosmic rays near the horizon, the average energy 1318 of the remaining cosmic rays increases. The sample dip also contributes to 1319 a change in the effective attenuation length and shielding, as described in 1320 Dunne et al. (1999). 1321

$$I(\theta) = I_0 \sin^m \theta, \tag{B.2}$$

where I is the particle beam intensity and θ is the inclination angle from 1322 the vertical. The exponent m, which is dependent on both energy and particle 1323 type, has been experimentally fit in several papers with significant differences 1324 in the results, as discussed in Dunai (2010). The most commonly cited value 1325 is 2.3 ± 0.5 from Nishiizumi et al. (1989), although Lal (1958) also gives a 1326 value of 2.3. However, Conversi and Rothwell (1954), cited by Lal, give a 1327 value of 2.1 ± 0.3 for 60 MeV nucleons (2.6 ± 0.2 for 750 MeV). In other 1328 studies, values of 2.5 \pm 0.5 to 3.0 \pm 0.5 (Barford and Davis, 1952), 3.5 \pm 1329 1.2 (Heidbreder et al., 1971), and 2.65 (Masarik et al., 2000) have also been 1330 provided. Using Sato's PARMA model, the exponent is ca. 2.8 for neutrons 1331 (calculated for neutrons >100 MeV, at solar minimum, 200 m altitude, R_c 1332 = 0 GV). The corresponding result for protons of similar energies is on the 1333 order of 3-3.5. Comparisons between this value and those reported in other 1334

studies must be considered with carefully because the energy ranges of the measurements are potentially quite different. Muons follow a similar trend, although the exponent in this case is 2 (Eidelman, 2004) or about 3.5 using the PARMA model. CRONUScalc does not use this formulation, however, and calculates the apparent attenuation length by interpolating from the values in Table 4, which were obtained using the spreadsheet published with Sato et al. (2008) and the adjustments described in the text.

The effective attenuation length depends on the sample location as well as 1342 the dip and shielding. The values for real samples vary from approximately 1343 150-190 g/cm², which overlaps with the measured values for the apparent 1344 attenuation length, Λ_f , for fast nucleonic particles, which range from 121 1345 to $>170 \text{ g/cm}^2$ (Dunai, 2000). Some recent studies for Be use a value of 1346 177 (Farber et al., 2008; Schimmelpfennig et al., 2008). In addition to the 1347 dependence of the effective attenuation length on topographic shielding, it 1348 also depends on site cutoff rigidity and elevation, due to the dependence 1349 of the particle penetration length on particle energy. Samples from lower 1350 elevation and lower latitude will have longer attenuation lengths because of 1351 the increasing hardness of the neutron energy spectrum. 1352

The attenuation-length model in CRONUScalc is based on atmospheric attenuation lengths calculated from the PARMA model of Sato et al. (2008). These are adjusted upward by 11.1% to account for systematic differences between atmospheric and lithospheric attenuation. See Section 2.4 for the complete derivation of the CRONUScalc attenuation length model. The data for the comparison of synthetic sample ages using different attenuation length models are presented here (B.6).

1360 Appendix B.2. Epithermal Neutrons

Low-energy cosmogenic nuclide production, including that from thermal 1361 and epithermal neutrons, does not follow a simple exponential pattern with 1362 depth due to the atmosphere-ground interface effects. Due to the large cross-1363 section of nitrogen for absorption of low-energy neutrons, the flux of cosmo-1364 genic thermal and epithermal neutrons in equilibrium with air is much less 1365 than that in equilibrium with rock. Low-energy neutrons produced in the 1366 upper ~ 50 cm of rock therefore tend to diffuse upward out of the rock and 1367 into the atmosphere, resulting in a reduction of the flux as the rock surface 1368 is approached (see Figure B.6). Phillips et al. (2001) analytically solved the 1369 neutron-flux differential equation across the land/atmosphere interface to 1370

		153 g/cm^2	170 g/cm^2
Erosion	Be conc	Age	Age
(mm/kyr)	(at/g)	(ka)	(ka)
0	73513	20.0	20.0
0	360327	100.0	100.0
0	1029138	300.0	300.0
1	72154	20.0	20.0
1	328482	100.0	99.1
1	789962	300.0	290.8
2.5	70156	20.0	19.9
2.5	287378	100.0	97.6
2.5	558968	300.0	272.2
5	66974	20.0	19.8
5	233598	100.0	94.7
5	354648	300.0	230.2
10	61176	20.0	19.6
10	163166	100.0	86.9
10	195499	300.0	156.6

Table B.6: Summary of comparison of exposure ages using two different attenuation lengths for a variety of erosion rates. Sample parameters assumed in the calculations for CRONUScalc were: 40 deg latitude, 0 deg longitude, 0 m elevation, thickness of 0.001 cm, shielding of 1, density of 2.65 g/cm³, collection date of 2014 AD.

obtain the equations below. These, in turn, were combined with the formulation for muon-induced neutron production by Gosse and Phillips (2001)
to obtain the complete equations for epithermal production.

The general form for the equation for the production of the cosmogenic nuclide of interest from epithermal neutrons is shown in Equation B.3. The epithermal neutron attenuation length ($\Lambda_{eth,ss}$, Equation B.4) accounts for both moderation and absorption of epithermal neutrons and parameterizes the effective depth of penetration of the epithermal neutron flux (Gosse & Phillips, 2001).

$$P_{eth,ss,m} = \frac{f_{eth,ss,m}}{\Lambda_{eth,ss}} \Phi_{eth,ss,total}(Z)(1 - p(E_{th})_{ss}), \tag{B.3}$$

where $f_{eth,ss,m}$ is the fraction of epithermal neutrons absorbed that are taken up by target element k to produce nuclide m (eqn E.15); $\Phi_{eth,ss,total}$ is the epithermal neutron flux (eqn B.8).

$$\Lambda_{eth,ss} = [\bar{\xi}_{bulk}(I_{eff,ss} + \Sigma_{sc,ss})]^{-1} = \Sigma_{eth,ss}^{-1}, \tag{B.4}$$

where ξ_{bulk} is the average macroscopic log decrement energy loss per neutron collision in the subsurface (eqn E.28); $I_{eff,ss}$ is the macroscopic resonance integral for absorption of epithermal neutrons in the subsurface (eqn E.16); $\Sigma_{sc,ss}$ is the macroscopic neutron scattering cross-section in the subsurface (Equation E.27).

The distribution of epithermal neutrons in the subsurface can be described by Equation B.5. The epithermal neutrons are assumed to be produced entirely from the moderation of both spallation and evaporation neutrons and are assumed to be in equilibrium with the high-energy flux (Gosse & Phillips, 2001).

$$D_{eth,ss} \frac{d^2 \Phi_{eth,ss}}{dZ^2} = \frac{\Phi_{eth,ss}}{\Lambda_{eth,ss}} - R_{eth,ss} P_f, \tag{B.5}$$

where the diffusion coefficient for epithermal neutrons in the subsurface $(D_{eth,ss})$ is defined in Equation E.9, with the corresponding parameter in the atmosphere, defined in Equation E.10; $R_{eth,ss}$ is the normalization factor for the epithermal neutron production rate (eqn E.25); P_f is the production rate of epithermal neutrons from fast neutrons, with the value at the surface represented as $P_f(0)$, which is a calibrated production rate parameter for 36 Cl.

The observed subsurface, epithermal neutron flux, $\Phi_{eth,ss}$, at a particular 1400 point is the balance of exponential production with depth against the loss of 1401 neutrons through diffusion at the rock/air interface as well as loss through 1402 absorption and moderation. The hypothetical epithermal neutron flux in the 1403 subsurface assuming that there is no boundary (all material is the same as 1404 the subsurface) is indicated by $\Phi_{eth,ss}^*$ and is given in Equation B.6. The 1405 difference between the hypothetical equilibrium flux in the air $(\Phi_{eth,a}^*)$ and 1406 the hypothetical equilibrium flux in the subsurface $(\Phi_{eth,ss}^*)$ is $\Delta \Phi_{eth,ss}^*$. The 1407 physical cause of the difference between the fluxes (no interface vs interface) 1408 is the effect of diffusion. This is shown mathematically in Equation B.7 and 1409 graphically in Figure B.6. 1410

$$\Phi_{eth,ss}^*(Z) = P_f(0) \frac{R_{eth,ss}}{\Sigma_{eth,ss} - D_{eth,ss} / \Lambda_{f,e}^2}$$
(B.6)

$$\Delta \Phi_{eth,ss}^* = \Phi_{eth,a}^* - \Phi_{eth,ss}^* = -\Delta \Phi_{eth,a}^* \tag{B.7}$$

For the purposes of this initial solution, photodisintegration and other neutron-producing interactions are neglected. The diffusion-like behavior of low-energy neutrons means that the diffusion equation, in addition to the production equations, must be solved in order to predict the appropriate amount of cosmogenic nuclide from this type of production. The solution to the coupled differential equations (originally solved in Phillips et al. (2001)) yields Equation B.8.

$$\Phi_{eth,ss} = S_T S_{el,s} \Phi^*_{eth,ss} exp(-\frac{Z}{\Lambda_{f,e}}) + (F\Delta\Phi)^*_{eth,ss} exp(-\frac{|Z|}{L_{eth,ss}}), \qquad (B.8)$$

where the physical meaning of the $(F\Delta\Phi)^*_{eth,ss}$ term (Equation B.9) is 1418 the difference between the actual observed flux (with an interface present) 1419 and the flux that would be observed at the surface (Z=0) if all materials 1420 were the same as those in the subsurface. This can also be formulated for 1421 the air, as seen in Equation E.12. $L_{eth,ss}$ (eqn E.19) is the diffusion length 1422 for epithermal neutrons in the subsurface and the corresponding parameter 1423 in the atmosphere, $L_{eth,a}$, is shown in Equation E.20. $S_{el,s}$ is the scaling 1424 factor for spallation reactions, as the flux is derived from the high-energy 1425 component. 1426

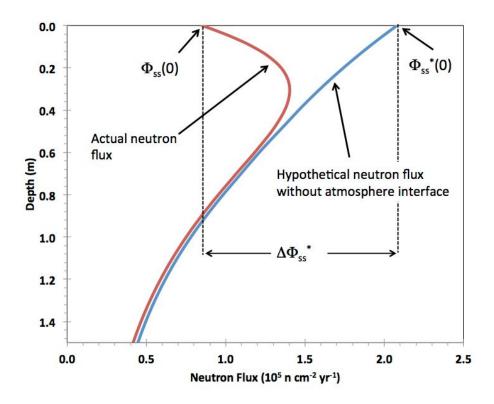


Figure B.6: Thermal-neutron flux profile calculated for standard granite from Fabryka-Martin (1988), illustrating the physical meaning of the terms $\Phi_{ss}(0)$, $\Phi_{ss}^*(0)$, and $\Delta \Phi_{ss}^*$. The physical meaning is equivalent for both thermal and epithermal neutron fluxes.

$$(F\Delta\Phi)_{eth,ss}^* = \frac{\Delta\Phi_{eth,ss}^* D_{eth,a}/L_{eth,a} - \Delta\Phi_{eth,a}^{**} D_{eth,ss}/\Lambda_{f,e}}{D_{eth,a}/L_{eth,a} + D_{eth,ss}/L_{eth,ss}},$$
(B.9)

1427 where:

$$\Delta \Phi_{eth,a}^{**} = \Delta \Phi_{eth,ss}^{*} - \frac{D_{eth,a}}{D_{eth,ss}} \Phi_{eth,a}^{*}$$
(B.10)

1428 Appendix B.3. Thermal Neutrons

Cosmogenic thermal neutrons are produced as a result of the moderation 1429 of cosmogenic epithermal neutrons (in other words, cosmogenic epithermal 1430 neutrons lose energy through collisions with atoms in the subsurface and end 1431 up as thermal neutrons, having only the ambient energy imparted by ordi-1432 nary thermal vibration). The source term for thermal-neutron production is 1433 therefore the epithermal neutron distribution with depth. The production 1434 equations for thermal neutron pathways are analogous to those for epither-1435 mal neutrons. In the general production equation, the form is very similar 1436 with many parameters being replaced by ones specific for thermal neutrons 1437 instead of epithermal neutrons. The production rate $(P_{th,m})$ for thermal 1438 neutrons is shown in Equation B.11 (Gosse & Phillips, 2001). 1439

$$P_{th,ss,m} = \frac{f_{th,ss,m}}{\Lambda_{th,ss}} \Phi_{th,ss,total}(Z), \qquad (B.11)$$

where $f_{th,ss,m}$ is the fraction of absorbed thermal neutrons that are taken up by element k to produce nuclide m (eqn E.32); $\Phi_{th,ss,total}$ is the thermal neutron flux (eqn B.13); and $\Lambda_{th,ss}$ is the effective thermal neutron attenuation length (Equation E.33).

The distribution for thermal neutrons in the subsurface (Equation B.12) is also similar to that of epithermal neutrons, except that the thermal neutron source is assumed to be only neutrons moderated from the epithermal energy range.

$$D_{th,ss} \frac{d^2 \Phi_{th,ss}}{dZ^2} = \frac{\Phi_{th,ss}}{\Lambda_{th,ss}} - R_{th,ss} \frac{p(E_{th})_a}{\Lambda_{eth,ss}} [\Phi^*_{eth,ss} exp(-\frac{Z}{\Lambda_{f,e}}) + (F\Delta\Phi)^*_{eth,ss} exp(-\frac{|Z|}{L_{eth,ss}})], \quad (B.12)$$

where $D_{th,ss}$ (Equation E.29) is the diffusion coefficient for thermal neutrons; $R_{th,ss}$ is the ratio of thermal neutron production in the rock to that in the atmosphere (Equation E.38). The term $p(E_{th})_a$ is the resonance escape probability of a neutron from the epithermal energy range in the atmosphere (Equation E.24), with the corresponding subsurface term shown in Equation E.23.

The equations described above can be solved for the thermal neutron flux using the same boundary conditions assumed in the epithermal problem. The thermal neutron flux, discounting muon-induced neutrons, is shown in Equation B.13.

$$\Phi_{th,ss} = S_T S_{el,s} \Phi_{th,ss}^* exp(-\frac{Z}{\Lambda_{f,e}}) + (\mathscr{F}\Delta\Phi)_{eth,ss}^* exp(-\frac{|Z|}{L_{eth,ss}}) + (\mathscr{F}\Delta\Phi)_{th,ss}^* exp(-\frac{|Z|}{L_{th,ss}})$$
(B.13)

1458 where:

$$\Phi_{th,ss}^* = \frac{p(E_{th})_a R_{th,ss} \Phi_{eth,ss}^*}{\Lambda_{eth,ss} (\Sigma_{th,ss} - \frac{D_{th,ss}}{\Lambda_{eth,ss}^2})}$$
(B.14)

$$(\mathscr{F}\Delta\Phi)^*_{eth,ss} = \frac{p(E_{th})_a R_{th,ss} (F\Delta\Phi)^*_{eth,ss}}{\Lambda_{eth,ss} (\Sigma_{th,ss} - D_{th,ss}/L^2_{eth,ss})}$$
(B.15)

$$(\mathscr{F}\Delta\Phi)_{th,ss}^{*} = [D_{th,a}(\frac{\Phi_{th,a}^{*}}{\Lambda_{f,e}} - \frac{(\mathscr{F}\Delta\Phi)_{eth,a}^{*}}{L_{eth,a}}) - D_{th}(\frac{\Phi_{th,ss}^{*}}{\Lambda_{f,e}} - \frac{(\mathscr{F}\Delta\Phi)_{eth,ss}^{*}}{L_{eth,ss}}) + \frac{D_{th,a}}{L_{th,a}}(\Delta\Phi_{th,ss}^{*} + \Delta(\mathscr{F}\Delta\Phi)_{eth,ss}^{*})]/(\frac{D_{th,ss}}{L_{th,ss}} + \frac{D_{th,a}}{L_{th,a}})$$
(B.16)

In Equation B.16, additional parameters of $\Delta \Phi_{th,ss}^*$ and $\Delta (\mathscr{F} \Delta \Phi)_{eth,ss}^*$ are described in Equations E.31 and E.4, respectively. $S_{el,s}$ is the scaling factor for spallation reactions.

1462 Appendix B.4. Muons

Muons are unstable, charged subatomic particles with a mass of about 207 times that of an electron and a mean life of about 2.2 s. They are produced by

decay of charged pions and K mesons, which in turn result from the interac-1465 tion of high-energy cosmic-ray protons with matter in the atmosphere and the 1466 solid earth (Lal & Peters, 1967). Cosmic-ray muons possess a wide spectrum 1467 of energies and the nature of their interaction with atomic nuclei depends 1468 strongly on energy. Lower energy ("slow") muons can participate in muon-1469 capture reactions. The muon-capture reactions can release neutrons that 1470 later participate in neutron-capture reactions. Higher energy ("fast") muons 1471 can produce nuclear transmutations through inelastic scattering reactions, 1472 including both primary scattering reactions by the muons themselves and 1473 secondary ones by hadrons and photons released as a result of the primary 1474 muon interaction. The hadrons released include neutrons, and neutrons are 1475 also produced by gamma rays from muon-induced bremsstrahlung reactions 1476 (emission of electromagnetic radiation from an atomic nucleus stimulated 1477 by deflection of an energetic charged particle passing close to the nucleus). 1478 The energetic bremsstrahlung gamma rays can produce neutrons if they par-1479 ticipate in (γ, n) reactions with the nuclei of other atoms. These neutrons 1480 also can later participate in neutron-absorption reactions. Muons can thus 1481 induce the production of the nuclides modeled by CRONUScalc by means 1482 of a wide variety of reactions (Charalambus, 1971; Fabryka-Martin, 1988; 1483 Gaisser, 1990; Heisinger et al., 2002a,b). 1484

Although the cosmic-ray muon flux is larger than the hadron flux at the 1485 earth's surface, the rate of interaction is much lower. The muonic component 1486 of the cosmic radiation is thus much more penetrating than the hadronic 1487 component. The slow component of the muon flux tends to be lost more 1488 rapidly due to muon-capture reactions and thus, although the magnitude 1489 of the flux decreases with depth below the earth's surface, the average en-1490 ergy increases. Whereas the hadron flux is important for cosmogenic-nuclide 1491 production down to about $1,000 \text{ g cm}^{-2}$, the slow-muon flux is important 1492 down to about $2,500 \text{ g cm}^{-2}$ and nuclides produced by the fast-muon flux 1493 are detectable down to $10,000 \text{ g cm}^{-2}$. The penetration of muons producing 1494 cosmogenic nuclides is often characterized by an attenuation length employed 1495 in an exponential depth-dependent equation (e.g., Braucher et al. 2013), but 1496 due to the continuous hardening of the muon energy spectrum with depth, 1497 the muon flux does not have an exponential dependence on depth and the 1498 apparent attenuation length will increase as the depth of the measurements 1499 increases. 1500

¹⁵⁰¹ Muon contributions to total production are typically small at the surface, ¹⁵⁰² but become predominant at depths exceeding $\sim 1,000$ g cm⁻². Accurate calculation of muogenic production is thus important for sampling sites with
large erosion rates or depth profiles. Accurate muon production formulations
are also necessary for depth profiles deeper than about 3 m (Stone et al., 1998;
Granger & Smith, 2000), where the simultaneous evaluation of spallogenic
and muogenic fractions offers the opportunity to uniquely determine both
age and erosion rate (Braucher et al., 2009).

The original CRONUS-Earth calculator of Balco et al. (2008) imple-1509 mented the Heisinger et al. (2002a,b) muon model as an improvement over 1510 the more empirical formulation in Stone et al. (1998). However, soon after 1511 Heisinger's publications, Braucher et al. (2003) used a deep core to provide 1512 evidence that the parameters specified by Heisinger et al. (2002a,b) overes-1513 timated actual ¹⁰Be production by fast muons by approximately a factor of 1514 two. This was supported by additional profile data measured by Braucher 1515 et al. (2011) and by Kim & Englert (2004), as well as reanalysis of previ-1516 ously published deep profile data by Braucher et al. (2013). This result has 1517 been confirmed by a deep profile drilled in a very low-erosion environment at 1518 Beacons Hills, Antarctica, by the CRONUS-Earth Project (Fig 1 in Phillips 1519 et al., 2015). CRONUScalc employs the Heisinger formulation for calculation 1520 of the subsurface muon flux for all nuclides, but uses calibrated production 1521 parameters obtained by fitting to the Beacon Heights profile data for ^{10}Be 1522 and ^{26}Al . 1523

The muon-flux model of Sato et al. (2008), combined with the previous Heisinger muon production equations and CRONUS-Earth data, has been employed to formulate a new hybrid muon production model that combines aspects of each of these models. The CRONUScalc muon code has been constructed by N. Lifton by modifying the Heisinger muon implementation described in Balco et al. (2008) and supplemental materials. The procedure followed by the code is summarized here along with the relevant equations.

The muon flux at the surface is scaled based on the Lifton-Sato-Dunai 1531 model (Lifton et al., 2014). The scaling is done by computing the omnidirec-1532 tional muon flux as a function of energy at the site and at a reference location 1533 (SLHL) and dividing the site flux by the reference flux. The CRONUScalc 1534 muon module initially calculates the omnidirectional muon flux at SLHL and 1535 at the site of interest using the PARMA-based equations (Sato et al., 2008). 1536 Energy-dependent scaling factors are then calculated as the ratio of site flux 1537 to that at SLHL for each energy interval. A single scaling factor is similarly 1538 calculated for the stopping muon flux at the surface - we define stopping 1539 muons as those that have ranges less than 10 g/cm^2 (equivalent to ca. 40 1540

¹⁵⁴¹ MeV energy) (Groom et al., 2001). The vertical muon flux at SLHL is calcu-¹⁵⁴² lated using the equation presented in Equation 3 in Heisinger et al. (2002a) ¹⁵⁴³ and shown here as Equation B.17.

$$\phi_{\nu,0}(\mathbf{Z}) = \frac{a}{(\mathbf{Z}+b)([\mathbf{Z}+1000]^{1.66}+c)}e^{d\mathbf{Z}},\tag{B.17}$$

where the coefficients are as follows: $a = 5.401 \times 10^7$, b = 21000, c = 15451.567 × 10⁵, $d = -5.5 \times 10^{-6}$. This formulation is only valid down to 200,000 g/cm² (~ 750 m depth) and the alternative formulation provided in Balco (2008 supplemental material - Eqns. 49-51) should be used for deeper applitations.

The stopping rate of the vertically incident muons $(R_{\nu,0}(Z))$ is calculated by equivalence to the range spectrum of the muons at the surface. Equation B.18 shows this calculation.

$$R_{\nu,0}(Z) = \frac{d}{dz}(\phi_{\nu,0}(Z)) = -5.401 \times 10^7 \left[\frac{bc\frac{da}{dz} - a\left(\frac{db}{dz}c + \frac{dc}{dz}b\right)}{b^2 c^2}\right], \qquad (B.18)$$

where $a = e^{-5.5 \times 10^{-6}} z$; b = (z + 21000); $c = (z + 1000)^{1.66} + 1.567 \times 10^5$; $\frac{da}{dz} = -5.5 \times 10^{-6} e^{-5.5 \times 10^{-6} z}$; $\frac{db}{dz} = 1$; $\frac{dc}{dz} = 1.66(z + 1000)^{0.66}$ (Balco et al., 1554 2008; Heisinger et al., 2002b).

The vertical muon flux as a function of depth $(\phi_{\nu}(\mathbf{Z}))$ at the site is found by numerically integrating the stopping rate from infinite depth to the depth of interest (Equation B.19).

$$\phi_{\nu}(\mathbf{Z}) = \int_{\mathbf{Z}}^{\infty} R_{\nu}(x) dx \tag{B.19}$$

The vertical muon flux is converted to total muon flux at the site following Heisinger et al. (2002b) (Equation B.20), but modified to use depth units of g/cm^2 by Balco et al. (2008), as shown in Equation B.21.

$$\phi(\mathbf{Z}, \theta) = \phi_{\nu}(\mathbf{Z}) cos^{n(\mathbf{Z})} \theta \tag{B.20}$$

$$n(\mathbf{Z}) = 3.21 - 0.297 \ln(\frac{\mathbf{Z}}{100} + 42) + \mathbf{Z} * 1.21 \times 10^{-5}$$
 (B.21)

The total muon flux at a given depth $(\phi_{\mu}(\mathbf{Z}))$ is given by Equation B.22.

$$\phi_{\mu}(\mathbf{Z}) = \frac{2\pi}{n(\mathbf{Z} + \delta \mathbf{Z}) + 1} \phi_{\nu}(\mathbf{Z}) \tag{B.22}$$

where δZ is the difference between the site of interest and sea level in units 1562 of g cm⁻². The total muon stopping rate as a function of depth (R(Z)) can 1563 be calculated by Equation B.23, following Balco (2007). The total stopping 1564 rate in muons $g^{-1} s^{-1}$ is converted to the negative muon stopping rate by 1565 accounting for the percentage of all stopped muons that are negative stopped 1566 muons (44%) and then converting the units to muons g^{-1} yr⁻¹. In both flux 1567 calculations (total muon flux and stopped negative muon flux) the values are 1568 calculated so that the flux is a positive value. 1569

$$R(Z) = \frac{2\pi}{n(Z + \delta Z) + 1} R_{\nu}(Z) - \phi_{\nu}(Z)(-2\pi) *$$
$$(n(Z + \delta Z) + 1)^{2} [\frac{-0.297 \times 10^{-2}}{\frac{Z + \delta Z}{100} + 42} + 1.21 \times 10^{-5}]$$
(B.23)

1570 Appendix B.4.1. Fast Muon Production

¹⁵⁷¹ Production from fast muons as a function of depth is calculated using the ¹⁵⁷² formulas in Heisinger et al. (2002b), with the general production described ¹⁵⁷³ by Equation B.24.

$$P_{\mu,fast} = S_T \phi_{\mu,total}(\mathbf{Z})\beta(\mathbf{Z})(\bar{\mathbf{E}}(\mathbf{Z}))^{\alpha}\sigma_0 \mathbf{N}_{t,i}$$
(B.24)

¹⁵⁷⁴ Where the factor $\beta(Z)$ is a function of the mean total muon energy and ¹⁵⁷⁵ is shown in Equation B.25 and \overline{E} is defined as the mean muon energy at a ¹⁵⁷⁶ given depth Z and is shown in Equation B.26. N_{t,i} is the number density of ¹⁵⁷⁷ the atoms in the target element (in units of at/g). This value is a constant ¹⁵⁷⁸ for each nuclide unless the composition of the target changes, as it does for ¹⁵⁷⁹ ³⁶Cl.

$$\beta(Z) = 0.846 - 0.015 \log(\frac{Z}{100} + 1) + 0.003139 (\log(\frac{Z}{100} + 1)^2)$$
(B.25)

$$\bar{E}(Z) = 7.6 + 321.7 * (1 - \exp(-8.059 \times 10^{-6}Z)) + 50.7(1 - \exp(-5.05 \times 10^{-7}Z))$$
(B.26)

In Heisinger et al. (2002b), the experimentally-determined value of σ_{190} 1580 (i.e., the energy-dependent reaction cross-section measured at 190 GeV) was 1581 used to calculate the value for σ_0 (the cross-section at 1 GeV) using Equation 1582 B.27, which can then be used to calculate the production from muons for a 1583 particular nuclide, as shown in Equation B.24. Alpha (α) is an only weakly 1584 energy-dependent coefficient that parameterizes the energy dependence of 1585 the cross-section on muon energy, with low values indicating a weaker de-1586 pendence and higher values a stronger dependence. Based on a survey of the 1587 literature, Heisinger et al. (2002b) adopted a value of $\alpha \approx 0.75$, but their 1588 own experimental evidence supported a value of about 0.93. 26 Al/ 10 Be ra-1589 tios measured by Kim & Englert (2004) in a profile down to 50,000 g/cm^{-2} 1590 depth are higher than predicted by Heisinger et al. (2002b), supporting a 1591 larger value of α . Experimental results are permissive of values between 0.75 1592 and >1.0 (Heisinger et al., 2002b) so the CRONUS-Earth Project chose a 1593 value of $\alpha = 1.0$. By assuming that α equals one, β will also be equal to 1594 one. Heisinger et al. (2002b) reported experimentally determined values of 1595 σ_{190} for many of the nuclides of interest including ²⁶Al, ¹⁰Be, ¹⁴C, and ³⁶Cl 1596 from calcium. Note that there is no value for ³⁶Cl from potassium. However, 1597 application of these values in many cases produced calculated concentrations 1598 in excess of what has been measured in deep profiles (Braucher et al., 2011, 1599 2013). 1600

1601 Rather than using parameters estimated from laboratory muon irradiations, the CRONUS-Earth Project has adopted values calibrated from nuclide-1602 concentration profiles at carefully selected sites whenever possible (Fig 1 in 1603 Phillips et al., 2015; Borchers et al., 2015; Marrero, 2012). σ_0 was selected as 1604 the calibration parameter for the production of nuclides by muon reactions, 1605 as discussed in Marrero (2012). This was done mainly for two reasons. First, 1606 σ_0 is the only nuclide-dependent parameter in the fast production equation, 1607 so it is the logical choice. Second, by directly calibrating the σ_0 parameter, 1608 any dependence on the accuracy of the conversion from σ_{190} to σ_0 is elim-1609 inated. Equation B.27 was used to calculate initial starting parameters for 1610 the σ_0 calibration. 1611

$$\sigma(E) = \sigma_0 E^{\alpha},\tag{B.27}$$

where E is the muon energy in GeV.

¹⁶¹³ Appendix B.4.2. Slow Negative Muon Capture Production

Nuclide production by slow negative muon capture $(P_{\mu-})$ is described by 1614 Equation B.28, originally from Charalambus (1971) and discussed in detail 1615 for 36 Cl by Stone et al. (1998). The production rate depends on the stopping 1616 rate of negative muons $(\phi_{\mu-})$ as well as the nuclide-dependent factors $(f_{i,C})$ 1617 $f_{i,D}, f_i^*$). $f_{i,D}$ is the fraction of muons stopped by element k and absorbed 1618 by the nucleus before decay of the muon. $f_{i,C}$, the compound factor, rep-1619 resents the fraction of the muons that are captured by a target element (as 1620 opposed to the other elements present) within the bulk rock. The formula 1621 for the compound factor is taken from Charalambus (1971) and the values 1622 are consistent with those used by Heisinger et al. (2002a). In the case of 1623 nuclides measured in a rock of uniform composition, the compound factor 1624 is a constant parameter; however, for cases where the lithology, and hence 1625 the chemical composition, is spatially variable, the compound factor will also 1626 vary. The value for $f_{i,C}$ can be calculated using the formula in Equation E.40 1627 and the values in von Egidy & Hartmann (1982). 1628

$$P_{\mu-} = S_T \phi_{\mu-}(Z) f_{i,C} f_{i,D} f_i^* \tag{B.28}$$

The remaining parameter, f_i^* , the particle emission channel probability, 1629 is the probability that the excited nucleus of the target atom will emit the 1630 proper particle to result in transformation to the nuclide of interest. Heisinger 1631 et al. (2002a) report experimentally determined f_i^* values for the production 1632 of ²⁶Al (from Si), ¹⁰Be and ¹⁴C (from O), and ³⁶Cl (from K and Ca). As for 1633 the fast muon production reactions, the parameters of Heisinger et al. (2002a) 1634 tend to overestimate nuclide concentrations measured in depth profiles (Fig 1635 1 in Phillips et al., 2015; Braucher et al., 2011), so f_i^* for ¹⁰Be, ²⁶Al, ³⁶Cl 1636 were calibrated by fitting to the measured CRONUS-Earth profiles (Phillips 1637 et al., 2015; Borchers et al., 2015; Marrero, 2012). 1638

1639 Appendix B.4.3. Muon-induced Neutrons

As muons react with atomic nuclei in the subsurface, neutrons are released. These can, in turn, react with other nuclei to produce cosmogenic nuclides of interest. The principal reactions responsible for the production of neutrons are muon capture and (γ, n) reactions resulting from interaction of high-energy gamma from muon-induced bremsstrahlung reactions with the nuclei of atoms in the subsurface. In order to calculate the nuclide production due to these muonic neutrons, it is necessary to know the muon flux as

a function of depth. Due to the capabilities of the muon model, the quan-1647 tification of fluxes has improved and they need no longer be approximated 1648 using a simple exponential equation (Stone et al., 1998), as was previously 1649 done in most models, including CHLOE (Phillips & Plummer, 1996) and the 1650 Schimmelpfennig et al. (2009) calculator, but instead are directly calculated 1651 using Equation B.29. The muon module, described in the previous section, is 1652 used to calculate the negative muon stopping rate $(\phi_{\mu-}(Z))$ and total muon 1653 flux $(\phi_{\mu f}(Z))$ terms at a given depth. 1654

$$P_{n,\mu}(Z) = Y_s \phi_{\mu-}(Z) + 5.8 \times 10^{-6} \phi_{\mu f}(Z), \qquad (B.29)$$

where Y_s is the average neutron yield per stopped negative muon (Fabryka-Martin, 1988).

Near the atmosphere/subsurface interface, muon-induced low-energy neu-1657 trons are assumed to follow the same distribution as the spallation-induced 1658 neutrons. Although the muon-induced neutron flux near the surface is not 1659 in equilibrium with the production rate due to diffusion, the diffusion is 1660 occurring based on the total concentration of neutrons at the surface. The 1661 dominant source of neutrons is spallation reactions, so the muons are assumed 1662 to follow the same pattern as the spallation-induced neutrons, leading to the 1663 incorporation of the muogenic neutrons into the epithermal neutron flux as 1664 shown in Equation B.30. 1665

$$\Phi_{\text{eth,ss}}(Z) = S_T S_{el,s} \Phi_{\text{eth,ss}}^* \exp\left(-\frac{Z}{\Lambda_{\text{f,e}}}\right) + (1 + R_{\mu}(0)R_{\text{eth,ss}})(F\Delta\Phi)_{\text{eth,ss}}^* \exp\left(-\frac{Z}{L_{\text{eth,ss}}}\right) + R_{\mu}(Z)\Phi_{\text{eth,ss}}^*$$
(B.30)

There are two different values used in the code for the muon-induced 1666 neutron factor, R_{μ} . $R_{\mu}(Z)$ is defined in Equation B.31. In the parts of the 1667 equation dealing with epithermal neutrons, the surface production rate of 1668 the muogenic neutrons $(R_{\mu}(0))$ is used because of the assumption that the 1669 production follows the same trend as the spallogenic neutrons. For the parts 1670 of the equation dependent on the attenuation at depth of the muon flux, the 1671 actual values for $R_{\mu}(Z)$ are calculated from the muon module and used in 1672 the equation. 1673

$$R_{\mu}(Z) = \frac{S_{el,\mu}P_{n,\mu}(Z)}{S_{el,eth}P_f(0)R_{eth}}$$
(B.31)

The code calculates the epithermal neutron flux and the subsequent cosmogenic nuclide production by combining the muon-induced neutrons with the original epithermal neutron production equation, Equation B.8, to produce the total epithermal neutron production for nuclide m, as shown in Equation B.32.

$$P_{\text{eth,ss,m}}(\mathbf{Z}) = \frac{f_{\text{eth,ss}}\Phi_{\text{eth,ss,total}}}{\Lambda_{\text{eth,ss}}} = \frac{f_{\text{eth,ss}}}{\Lambda_{\text{eth,ss}}} \left\{ \Phi_{\text{eth,ss}}^* \exp\left(-\frac{\mathbf{Z}}{\Lambda_{\text{f,e}}}\right) + (1 + R_{\mu}(0)R_{\text{eth,ss}})(F\Delta\Phi)_{\text{eth,ss}}^* \exp\left(-\frac{\mathbf{Z}}{L_{\text{eth,ss}}}\right) + R_{\mu}(\mathbf{Z})\Phi_{\text{eth,ss}}^* \right\}$$
(B.32)

Similar considerations for muon-induced neutrons must be made for the 1679 thermal energy range. Some of the epithermal-range neutrons produced by 1680 muon interactions lose enough energy to become thermal neutrons. The 1681 contribution of these muon-induced neutrons to the thermal flux is shown 1682 in Equation B.33. Once again, the muon-induced neutrons are assumed to 1683 follow the same trends as the spallogenic neutrons and are scaled appropri-1684 ately using only the surface value of $R'_{\mu}(Z)$ for the spallogenic parts of the 1685 equation. For the neutron-dependent term, the actual values for each depth 1686 are calculated. The final equation for the thermal neutron flux is shown in 1687 Equation B.34. 1688

$$R'_{\mu}(Z) = \frac{p(E_{th})_a}{p(E_{th})} R_{\mu}(Z)$$
(B.33)

$$\Phi_{th,ss,total} = S_T S_{el,s} \left\{ \Phi_{th,ss}^* \exp\left(-\frac{Z}{\Lambda_{f,e}}\right) + (1 + R'_{\mu}(0))(\Im \Delta \Phi)_{eth,ss}^* \exp\left(-\frac{Z}{L_{eth,ss}}\right) + (1 + R'_{\mu}(0)R_{th,ss})(\Im \Delta \Phi)_{th,ss}^* \exp\left(-\frac{Z}{L_{eth,ss}}\right) + R'_{\mu}(Z)\Phi_{th,ss}^* \right\}$$
(B.34)

The total production of cosmogenic nuclide k via thermal neutron pathways is described by combining Equation B.34 with Equation B.11 and yields an equation for total production with depth from thermal neutrons, shown in Equation B.35.

$$P_{\rm th,ss,m}(Z) = S_T S_{el,s} \frac{f_{\rm th} \Phi_{\rm th,ss,total}}{\Lambda_{\rm th,ss}} = \frac{f_{\rm th}}{\Lambda_{\rm th,ss}} \left\{ \Phi_{\rm th,ss}^* \exp\left(-\frac{Z}{\Lambda_{\rm f,e}}\right) + (1 + R'_{\mu}(0))(\Im\Delta\Phi)_{\rm eth,ss}^* \exp\left(-\frac{Z}{L_{\rm eth,ss}}\right) + (1 + R'_{\mu}(0)R_{\rm th})(\Im\Delta\Phi)_{\rm th,ss}^* \exp\left(-\frac{Z}{L_{\rm eth,ss}}\right) + R'_{\mu}(Z)\Phi_{\rm th,ss}^* \right\}$$
(B.35)

¹⁶⁹³ Appendix B.5. Radiogenic Production

"Radiogenic production" in this context refers to the generation of low-1694 energy neutrons by reactions related to the radioactive decay or spontaneous 1695 fission of U or Th, and the subsequent absorption of those neutrons to pro-1696 duce nuclides of interest, principally ³⁶Cl. The radiogenic low-energy neutron 1697 flux is assumed to be in equilibrium with the concentrations of uranium (U) 1698 and thorium (Th) in the rock. This component is quantified using measured 1699 concentrations of U and Th and the method described in Fabryka-Martin 1700 (1988), which is based on the formulations developed by Feige et al. (1968). 1701 The uranium and thorium α -decay chain members produce alpha particles 1702

 (α) as they decay. The alpha particles react with light nuclei in the rock matrix to produce low-energy neutrons. In turn, the neutrons can react with target elements in the rock in the same way that cosmogenically produced neutrons react to produce nuclides such as ³⁶Cl. The equations for calculating this contribution to the total nuclide inventory within a sample are provided in detail in Fabryka-Martin (1988) and are summarized here.

The radiogenic production of nuclides is given by Equation B.36. The 1709 elements with the maximum yield of neutrons due to alpha particle reaction 1710 are Be, B, F, and Li. However, due to the low concentration of these elements 1711 in most rocks, the largest concentrations of neutrons result from targets 1712 with larger matrix concentrations, such as Al, Si, Mg, O, and Na. The 1713 concentrations of both O and H are calculated from the oxide measurements 1714 performed on the other elements. 1715

$$P_r = P_{eth,r} f_{eth} + P_{th,r} f_{th} \tag{B.36}$$

where P_r is the total radiogenic production from all mechanisms in a particular sample; $P_{eth,r}$ is the total radiogenic epithermal neutron production (Equation B.37); $P_{th,r}$ is the total radiogenic thermal neutron production (Equation B.38).

Although the concentrations of the largest producers of neutrons are the 1720 most important elements to quantify, the remaining rock matrix composition 1721 must still be quantified in addition to the elements listed above so that all 1722 elements can be used to calculate the stopping power of the rock. In par-1723 ticular, there are several elements, such as boron and gadolinium, that have 1724 large thermal neutron absorption cross-sections, meaning that they have a 1725 large probability of absorbing neutrons (from both radiogenic and cosmo-1726 genic sources) (Fabryka-Martin, 1988). This decreases the actual amount 1727 of 36 Cl formed within the rock because these other elements intercept the 1728 neutrons prior to formation of 36 Cl. 1729

$$P_{eth,r} = (P_{n,\alpha} + P_{n,sf})(1 - p(E_{th})),$$
(B.37)

$$P_{th,r} = (P_{n,\alpha} + P_{n,sf})(p(E_{th})),$$
(B.38)

where $P_{n,\alpha}$ is the production rate of neutrons from alpha particles in neutrons g⁻¹ yr⁻¹ (Equation B.39). $P_{n,sf}$ is the neutron production rate due to the spontaneous fission of ²³⁸U and can be calculated as 0.429[U], where [U] is the concentration of uranium in ppm.

$$P_{n,\alpha} = X[U] + Y[Th], \tag{B.39}$$

where [Th] is the thorium concentration in ppm. X and Y are neutron production factors related to the light isotope composition of the rock matrix and are described in Equations B.40 and B.41, respectively. These were originally described by Feige et al. (1968).

$$X = \frac{\sum k S_k F_{k,bulk} Y_n^U}{\sum k S_k F_{k,bulk}},$$
(B.40)

$$Y = \frac{\sum k S_k F_{k,bulk} Y_n^{Th}}{\sum k S_k F_{k,bulk}},$$
(B.41)

where S_k is the mass stopping power of element k for α -particles of a given energy; Y_n^U and Y_n^{Th} are the neutron yields of element i per ppm U or Th in equilibrium; $F_{k,bulk}$ is the fractional abundance of element k in ppm in the bulk rock.

1742 Appendix C. Scaling

Cosmogenic nuclide scaling applies the physics governing the modulation 1743 of the cosmic-ray flux by atmospheric mass and the terrestrial and solar 1744 magnetic fields to provide production rates as a function of location and 1745 exposure time. Numerous scaling frameworks have been proposed in order 1746 to correct for latitude, elevation, atmospheric pressure anomalies, dipole and 1747 non-dipole geomagnetic field changes, and solar modulation. For all the 1748 details of the scaling frameworks themselves, please see the original papers 1749 (cited in Table 2); for details of implementation, please see the descriptions 1750 in Balco et al. (2008) and Lifton et al. (2014). 1751

The fundamental correction for elevation and latitude is the key part 1752 of each scaling framework. The first scaling framework to implement this 1753 was Lal (1991). Eventually, Stone (2000) reformulated the original equa-1754 tions to take atmospheric pressure, instead of elevation, as an input. This 1755 is still a commonly cited scaling framework. However, the original, time-1756 independent Lal/Stone scaling (abbreviated as St in the code) does not ac-1757 count for changes in production rate that we know are occurring through 1758 time due to fluctuations in the terrestrial and solar magnetic fields. 1759

The production rates of cosmogenic nuclides are a function of the magnetic field strength. As the geomagnetic field of the earth changed in the

past, production rates of the nuclides also changed. This fact has led to the 1762 development of scaling frameworks that incorporate the geomagnetic history 1763 of the earth. Dunai (2000, 2001a) (Du), Desilets et al. (2006b); Desilets & 1764 Zreda (2003) (De), and Lifton et al. (2005, 2008) (Li), and Lifton-Sato-Dunai 1765 (Lifton et al., 2014) (LSD, herein labeled Sf) have all implemented different 1766 models to incorporate the changing magnetic field. Following Balco et al. 1767 (2008), we have also provided the time-dependent Lal scaling (Lm, including 1768 dipole geomagnetic effects as approximated by Nishiizumi et al. (1989)) in 1769 order to differentiate between the geomagnetic effects and other differences 1770 in the models. 1771

In CRONUScalc, the geomagnetic history is consistent across all scaling frameworks and is summarized in Table 3. The Lifton et al. (2005) and the more recent model presented in Lifton et al. (2014) (LSD) also incorporate the effects of solar modulation into the scaling framework. For a discussion of the uncertainties associated with geomagnetic models, see Section 2.3.

In each scaling framework that incorporates geomagnetic effects, the ap-1777 propriate input is cutoff rigidity. However, each scaling framework imple-1778 ments the rigidity cutoff calculations differently (see Schimmelpfennig (2009) 1779 for a summary of these differences). See each original scaling reference for the 1780 equations for cutoff rigidity. In the program, most of the scaling frameworks 1781 are taken directly from Balco et al. (2008) and complete descriptions can be 1782 found in the original calculator supplemental material (Balco et al., 2008). 1783 For the Sa and Sf models, see the paper by Lifton et al. (2014). 1784

1785Appendix D. Comparison of CRONUScalc and Analytical Solu-1786tions for ³He

Synthetic ³He concentrations were predicted using the analytical solution 1787 presented in Dunai (2010) for samples undergoing both production and ero-1788 sion. The CRONUS-Earth calibrated production rate for ST scaling and the 1789 erosion rate (Table D.7, column 1) were used to predict the concentration 1790 (Table D.7, column 2). The resulting CRONUScale ages for ST scaling are 1791 presented in Table D.7, column 4. The differences in terms of % are provided 1792 in the final column of the table. Saturated samples were excluded from com-1793 parison. Samples were assumed to be located at SLHL, 0 degrees longitude, 1794 and have a density of 2.65 g/cm³, shielding of 1, thickness of 0.001 cm, an 1795 effective attenuation length of 136 g/cm^2 and were collected in 2014 AD. 1796

Erosion rate	Concentration	Actual age	Calc age	% Difference
(mm/kyr)	$(atoms g^{-1})$	(ka)	(ka)	(%)
0	118	0.001	0.001	4.6×10^{-7}
0	11821	0.1	0.100	2.0×10^{-7}
0	1182137	10	10.000	-4.9×10^{-7}
0	5910685	50	50.000	5.6×10^{-7}
0	11821370	100	100.000	5.6×10^{-7}
0	23642739	200	200.000	5.6×10^{-7}
0	47285479	400	400.000	5.6×10^{-7}
0	118213696	1000	1000.000	-7.5×10^{-7}
0	236427393	2000	2000.000	-7.5×10^{-7}
0	354641089	3000	3000.000	-9.9×10^{-7}
0	591068482	5000	5000.000	6.0×10^{-7}
0	827495875	7000	7000.000	8.5×10^{-7}
1	11820	0.1	0.100	2.0×10^{-7}
1	1170695	10	10.000	6.8×10^{-7}
1	5631906	50	50.000	5.6×10^{-7}
1	10741040	100	100.000	5.6×10^{-7}
1	19580586	200	200.000	5.6×10^{-7}
1	32842095	400	400.000	5.6×10^{-7}
1	52026941	1000	1000.000	7.5×10^{-7}
1	59441147	2000	2000.000	2.2×10^{-6}
1	60497723	3000	3000.000	8.9×10^{-6}
1	60669750	5000	5000.014	2.8×10^{-4}
1	60673244	7000	7000.662	9.5×10^{-3}
10	11810	0.1	0.100	11.7×10^{-5}
10	1074104	10	10.000	1.7×10^{-5}
10	3776908	50	50.000	2.7×10^{-5}
10	5202694	100	100.000	4.9×10^{-5}
10	5944115	200	200.000	2.0×10^{-4}
10	6064829	400	400.020	4.9×10^{-3}

Table D.7: Summary of comparison between analytical solutions for 3 He and CRONUScalc results over a range of erosion rates and exposure ages.

1797 Appendix E. Glossary of Terms and Equations

Term	Definition	
General parameters and subscripts		
bulk	In reference to the bulk rock composition of the sample	
	as opposed to the target (processed) material	
C_k	Mass concentration of element k [(g element k) (g material) ⁻¹]	
eth	Subscript used to denote the epithermal production path-	
	way, defined as neutrons with energies of 0.5 eV to 0.1	
	MeV.	
h	Atmospheric depth $[g \text{ cm}^{-2}]$	
k	Subscript used to denote a particular cosmogenic-	
	producing element	
m	Subscript used to denote a particular cosmogenic nuclide	
m	Molar concentration	
μ	Subscript used to denote the muon production pathway	
ρ	Density $[g \ cm^{-3}]$	
S	Subscript used to denote the spallation production path-	
	way	
target	In reference to the particular target fraction of the sam-	
	ple, specifically in the case of mineral separates	
th	Subscript used to denote the thermal production path-	
	way, defined as neutrons with energies of < 0.5 eV.	
Z	Ordinary linear distance [cm]	
Ζ	Mass depth below the surface $[g \text{ cm}^{-2}]$	
	$Z(z) = \int_0^z \rho(z) dz \tag{E.1}$	

Table E.8: Glossary of terms and equations

Cosmic Rays

С	Speed of light (see eqn A.1)
e	Particle charge (see eqn A.1)
p	Particle momentum (see eqn A.1)
	Continued on next page

Table E.8 – continued from previous page		
Term	Definition	
R	Rigidity of a particle (see eqn A.1)	
S_{el}	Latitude/Elevation scaling coefficient (see scaling scheme	
	section) [unitless]	
S_T	Topographic scaling coefficient describing the shielding	
	from surrounding topography [unitless]	
θ	Inclination angle from the horizontal [degrees]	
z_p	Vertical penetration depth $[g \text{ cm}^{-2}]$	
	$z_p = \Lambda_{f,p} cos\phi \tag{E.2}$	
Spallation		
$I(\theta)$	Intensity. See eqn B.2. I_0 is the intensity of I for a sample	
	with 0 dip.	
$\Lambda_{\mathrm{f,e}}$	Effective attenuation length for fast neutrons. See Ap-	
	pendix B.1.1.	
$P_{s,m}$	Production rate for spallation of nuclide m. See eqn 1.	
$P_{m,k}$	Production rate of nuclide m from target element k. See	
	eqn 1.	
θ	Sample dip/inclination as measured from the horizontal.	
	See eqn B.2.	
Epithermal P	roduction	
\bar{A}_{ss}	Average atomic weight of the bulk rock $[g mol^{-1}]$	
	$\bar{A}_{ss} = \frac{\sum_{k} A_k N_{k,ss,bulk}}{\sum_{k} N_{k,ss,bulk}} $ (E.3)	
$ar{A}_a$	Average atomic weight of the atmosphere; constant = $14.5 \text{ [g mol}^{-1}\text{]}$ (Phillips & Plummer, 1996)	
A_k	Atomic weight of element k $[g \text{ mol}^{-1}]$	
10	Continued on next page	

Table E.8 – continued from previous page

Term	Definition
$\Delta(\mathscr{F}\Delta\Phi)^*_{eth,ss}$	Difference between $\mathscr{F}\Delta\Phi)^*_{eth}$ in the atmosphere
, , , , , , , , , , , , , , , , , , , ,	$(\mathscr{F}\Delta\Phi)^*_{eth,a})$ and the subsurface $(\mathscr{F}\Delta\Phi)^*_{eth,ss})$
	$\Delta(\mathscr{F}\Delta\Phi)^*_{eth,ss} = (\mathscr{F}\Delta\Phi)^*_{eth,a} - (\mathscr{F}\Delta\Phi)^*_{eth,ss} \qquad (E.4)$
$\Delta \Phi^*_{eth,ss}$	Difference between the hypothetical equilibrium epither-
	mal neutron fluxes in atmosphere and rock [neutrons $cm^{-2} yr^{-1}$]
	$\Delta \Phi_{eth,ss}^* = \Phi_{eth,a}^* - \Phi_{eth,ss}^* \tag{E.5}$
	ein,ss ein,a ein,ss (°)
Δ .	Difference between the hundthatical equilibrium with a
$\Delta \Phi^*_{eth,a}$	Difference between the hypothetical equilibrium epither-
	mal neutron fluxes in atmosphere and rock [neutrons $cm^{-2} yr^{-1}$]
	$\Delta \Phi_{eth,a}^* = \Phi_{eth,ss}^* - \Phi_{eth,a}^* \tag{E.6}$
	$\Delta \Psi_{eth,a} - \Psi_{eth,ss} - \Psi_{eth,a} \tag{E.0}$
$\Delta \Phi_{eth,ss}^{**}$	Adjusted difference between the hypothetical equilibrium
	epithermal neutron fluxes in atmosphere and rock. [neutrons $cm^{-2} yr^{-1}$]
	\mathcal{D} .
	$\Delta \Phi_{eth,ss}^{**} = \Phi_{eth,a}^* - \frac{D_{eth,ss}}{D_{eth,ss}} \Phi_{eth,ss}^* $ (E.7)
	$ u_{eth,a}$
۸	
$\Delta \Phi_{eth,a}^{**}$	Adjusted difference between the hypothetical equilibrium
	epithermal neutron fluxes in atmosphere and rock. [neutrons $em^{-2} m^{-1}$]
	trons $\rm cm^{-2} \ yr^{-1}$]
	$D_{eth,a} D_{eth,a} (\Gamma \circ)$
	$\Delta \Phi_{eth,a}^{**} = \Phi_{eth,ss}^* - \frac{D_{eth,a}}{D_{eth,ss}} \Phi_{eth,a}^* $ (E.8)
	Continued on next page
	Pw80

Table E.8 – continued from previous page

	Definition
Term	
$D_{eth,ss}$	Diffusion coefficient for epithermal neutrons in subsurface $[g \text{ cm}^{-2}]$ $D_{eth,ss} = [3\Sigma_{sc}(1 - \frac{2}{3\bar{A}})]^{-1} (E.9)$
$D_{eth,a}$	Diffusion coefficient for epithermal neutrons in air [g cm ⁻²] can be calculated using values of $(\Sigma_{sc,a})=0.3773$ [cm ⁻² g ⁻¹] (Phillips & Plummer, 1996).
	$D_{eth,a} = [3\Sigma_{sc,a}(1 - \frac{2}{3\bar{A}})]^{-1} $ (E.10)
$(F\Delta\Phi)^*_{eth,ss}$	Difference between the epithermal neutron flux if there was no boundary $(\Phi_{eth,ss}^*)$ and the actual epithermal neu- tron flux at the atmosphere/subsurface interface (see equation B.9) [neutrons cm ⁻² yr ⁻¹] $(F\Delta\Phi)_{eth,ss}^* = \frac{\Delta\Phi_{eth,ss}^* D_{eth,a}/L_{eth,a} - \Delta\Phi_{eth,a}^{**} D_{eth,ss}/\Lambda_{f,e}}{D_{eth,a}/L_{eth,a} + D_{eth,ss}/L_{eth,ss}}$ (E.11)
$(F\Delta\Phi)^*_{eth,a}$	Difference between $\Phi_{eth,a}^*$ (the epithermal neutron flux in the atmosphere if there was no boundary) and $\Phi_{eth,a}$ (the actual epithermal neutron flux at the atmo- sphere/subsurface interface) [neutrons cm ⁻² yr ⁻¹] $(F\Delta\Phi)_{eth,a}^* = \frac{\Delta\Phi_{eth,a}^*D_{eth,ss}/L_{eth,ss} - \Delta\Phi_{eth,a}^{**}D_{eth,ss}/\Lambda_{f,e}}{D_{eth,a}/L_{eth,a} + D_{eth,ss}/L_{eth,ss}}$ (E.12)
	Continued on next page
	Commuted on next page

Table E.8 – continued from previous page

Term	Definition
$(\mathscr{F}\Delta\Phi)^*_{eth,ss}$	Describes the difference between $\Phi_{eth,ss}^*$ and the actual
()eth,ss	flux due to the shape of the epithermal neutron profile
	across the atmosphere/subsurface interface
	across the atmosphere/substituee interface
	$p(E_{th})_a R_{th,ss} (F\Delta\Phi)^*_{eth,ss}$ (E.12)
	$(\mathscr{F}\Delta\Phi)^*_{eth,ss} = \frac{p(E_{th})_a R_{th,ss} (F\Delta\Phi)^*_{eth,ss}}{\Lambda_{eth,ss} (\Sigma_{th,ss} - D_{th,ss}/L^2_{eth,ss})} (E.13)$
$(\mathscr{T} \Delta \Phi)^*$	Describes the difference between Φ^* and the actual
$(\mathscr{F}\Delta\Phi)^*_{eth,a}$	Describes the difference between $\Phi_{eth,a}^*$ and the actual
	flux due to the shape of the epithermal neutron profile
	across the atmosphere/subsurface interface
	$p(E_{th})_{a}R_{th}a(F\Delta\Phi)^{*}_{ah}$
	$(\mathscr{F}\Delta\Phi)_{eth,a}^* = \frac{p(E_{th})_a R_{th,a} (F\Delta\Phi)_{eth,a}^*}{\Lambda_{eth,a} (\Sigma_{th,a} - D_{th,a}/L_{eth,a}^2)} $ (E.14)
	$\Lambda_{eth,a}(\Delta_{th,a} - D_{th,a}/L_{eth,a})$
$f_{eth,m,ss}$	Fraction of total epithermal neutrons absorbed per unit
	mass of rock that react to produce nuclide m; composi-
	tionally dependent [unitless]
	N, I ,
	$f_{eth,m,ss} = \frac{N_{k,ss}I_{a,k}}{I_{eff}} \tag{E.15}$
	I_{eff}
$\Gamma_{eth,m,ss}$	Total rate of epithermal neutron absorption in subsurface
- ein,m,ss	[neutrons g^{-1} yr ⁻¹]
$I_{a,k}$	Dilute resonance integral for absorption of epithermal
- <i>a</i> , <i>κ</i>	neutrons by element k $[10^{-24}cm^{-2}]$
$I_{eff,ss}$	Effective/macroscopic resonance integral for absorption
-ejj,ss	of epithermal neutrons in subsurface $[cm^{-2}g^{-1}]$
	$I_{eff} = \sum I_{a,k} N_{k,bulk} \tag{E.16}$
	$I_{eff} = \sum_{k} I_{a,k} N_{k,bulk} \tag{E.16}$
	Continued on next page

Table E.8 – continued from previous page

Term	Definition
$\Lambda_{eth,ss}$	Effective epithermal neutron attenuation length in sub- surface, accounting for both absorption and moderation $[g \text{ cm}^{-2}]$
	$\Lambda_{eth,ss} = [\xi(I_{eff,ss} + \Sigma_{sc,ss})]^{-1} = \Sigma_{eth,ss}^{-1} $ (E.17)
$\Lambda_{eth,a}$	Effective epithermal neutron attenuation length in the atmosphere, accounting for both absorption and moder- ation $[g \text{ cm}^{-2}]$
	$\Lambda_{eth,a} = [\xi(I_{eff,a} + \Sigma_{sc,a})]^{-1} = \Sigma_{eth,a}^{-1} $ (E.18)
$L_{eth,ss}$	Diffusion length for epithermal neutrons in the subsurface $[g \text{ cm}^{-2}]$
	$L_{eth,ss} = (\sqrt{3\Sigma_{sc,ss}\Sigma_{eth,ss}})^{-1} $ (E.19)
$L_{eth,a}$	Diffusion length for epithermal neutrons in the air $[g cm^{-2}]$
	$L_{eth,a} = (\sqrt{3\Sigma_{sc,a}\Sigma_{eth,a}})^{-1} $ (E.20)
$N_{k,ss,bulk/target}$	Atomic concentration of element k in subsurface (target or bulk specified as additional subscript) $[at/g]$
$N_{k,a}$	Atomic concentration of element k in air $[at/g]$
$P_{eth,m}$	Production rate for epithermal production of nuclide m. See eqn B.28.
$\Phi_{eth,ss}(z)$	Epithermal neutron flux in subsurface [neutrons cm^{-2} yr^{-1}]
	Continued on next page

Table E.8 – continued from previous page

Term	Definition
$\Phi^*_{eth,ss}$	Epithermal neutron flux that would be observed at the
	land surface if the properties of the medium did not
	change (e.g. identical to the subsurface).) [neutrons
	$cm^{-2} yr^{-1}$] (See equation B.6)
	$\Phi_{eth,ss}^* = P_f(0) \frac{R_{eth,ss}}{\Sigma_{eth,ss} - \frac{D_{eth,ss}}{\Lambda_{f,e}^2}} $ (E.21)
$\Phi^*_{eth,a}$	Epithermal neutron flux that would be observed at the
	land surface if the properties of the medium did not
	change (e.g. atmosphere identical to the air).) [neutrons
	$cm^{-2} yr^{-1}$
	$\Phi_{etha}^* = \mathbf{P}_f(0) \frac{R_{eth,a}}{D} \tag{E.22}$
	$\Phi_{eth,a}^* = P_f(0) \frac{R_{eth,a}}{\Sigma_{eth,a} - \frac{D_{eth,a}}{\Lambda_c^2}} $ (E.22)
	$\Lambda_{f,e}$
$P_f(Z)$	Production rate of epithermal neutrons from fast sec-
-) (-)	ondary cosmogenic neutrons as a function of depth. [neu-
	trons (g air) ⁻¹ yr ⁻¹]
$P_f(0)$	is P_f at depth equal to 0 and is a calibrated production
1 f(0)	rate parameter. [neutrons (g air) ⁻¹ yr ⁻¹]
(F)	Resonance escape probability in the subsurface - proba-
$p(E_{th})_{ss}$	
	bility that a neutron will pass through the epithermal en-
	ergy range to the thermal range without being absorbed
	[unitless]
	$p(E_{th}) = exp[-\frac{I_{eff}}{\sum_{k} \xi_k N_{k,bulk} \sigma_{sc,k}}] $ (E.23)
	Continued on next page
Continued on next page	

Table E.8 – continued from previous page $% \left({{{\bf{F}}_{{\rm{B}}}} \right)$

Term	Definition
$p(E_{th})_a$	Resonance escape probability in the air - probability that
I (), u	a neutron will pass through the epithermal energy range
	to the thermal range without being absorbed [unitless].
	Value according to Phillips & Plummer (1996).
	$p(E_{th})_a = 0.56$ (E.24)
D .	Patie of anithermal neutron production in the reak to
$R_{eth,ss}$	Ratio of epithermal neutron production in the rock to
	that of the atmosphere [unitless]
	$\sqrt{\overline{A}}$
	$R_{eth,ss} = \sqrt{\frac{\bar{A}_{ss}}{A_a}} \tag{E.25}$
	$\bigvee A_a$
$R_{eth,a}$	Ratio of epithermal neutron production in the atmo-
	sphere to that of the atmosphere [unitless].
	$R_{eth,a} = \sqrt{\frac{\bar{A}_a}{A_a}} = 1 \tag{E.26}$
	$\bigvee A_a$
$\Sigma_{eth,ss}$	Effective epithermal loss cross-section in subsurface, by
	both absorption and energy moderation $[cm^2 g^{-1}]$
$\Sigma_{eth,a}$	Effective epithermal loss cross-section in air, by both ab-
<u> </u>	sorption and energy moderation $[\text{cm}^2 \text{ g}^{-1}]$
$\Sigma_{sc,ss}$	Macroscopic neutron scattering cross-section in subsur-
	face $[\rm cm^2 g^{-1}]$
	$\Sigma_{sc,ss} = \sum N_{k,bulk} \sigma_{sc,k} \tag{E.27}$
	$\Sigma_{sc,ss} = \sum_{k} N_{k,bulk} \sigma_{sc,k} \tag{E.27}$
	r.
$\Sigma_{sc,a}$	Macroscopic neutron scattering cross-section in air. Con-
sc,u	stant = $0.3773 [\text{cm}^2 \text{g}^{-1}]$
	Continued on next page

Table E.8 – continued from previous page

Table E.8 – continued from previous page	
Term	Definition
$\sigma_{sc,k}$	Neutron scattering cross-section for element k $[1 \times 10^{-24}]$
	$[\rm cm^2]$
$\overline{\xi}_{bulk}$	Macroscopic average log decrement neutron energy loss
	per collision for the bulk rock
	$\bar{\xi}_{bulk} = \frac{\sum_{k} \xi_k \sigma_{sc,k} N_{k,ss,bulk}}{\sum_{k} \sigma_{sc,k} N_{k,ss,bulk}} $ (E.28)
ξ_k	Average log decrement of energy loss for element k
Thermal Produ	iction

Table E.8 – continued from previous page

Thermal Produ	
$D_{th,ss}$	Diffusion coefficient for thermal neutrons in the subsur-
	face [unitless]
	$D_{th,ss} = [3\Sigma_{sc,ss}(1 - \frac{2}{3A_ss})]^{-1} $ (E.29)
$D_{th,a}$	Diffusion coefficient for thermal neutrons in the atmo- sphere [unitless]
	$D_{th,a} = [3\Sigma_{sc,a}(1 - \frac{2}{3A_a})]^{-1} $ (E.30)
$\Delta \Phi^*_{th,ss}$	Describes the difference between the hypothetical equi-
	librium thermal neutron flux in the air and the subsurface [neutrons $cm^{-2}yr^{-1}$]
	$\Delta \Phi_{th,ss}^* = \Phi_{th,a}^* - \Phi_{th,ss}^* \tag{E.31}$
	Continued on next page

Term	Definition	
-		
$f_{th,ss,m}$	Fraction of thermal neutrons absorbed per unit mass	
	by target k that react to form cosmogenic nuclide m	
	[unitless]	
	$\sigma_{th,k}N_{k,ss,target}$ (F. 22)	
	$f_{th,ss,m} = \frac{\sigma_{th,k} N_{k,ss,target}}{\Sigma_{th,ss}} $ (E.32)	
$(\mathscr{F}\Delta\Phi)^*_{th,ss}$	Describes the difference between $\Phi_{th,ss}^*$ and the actual	
,	flux due to the shape of the thermal neutron flux profile	
	across the interface. See equation B.16	
$\Lambda_{th,ss}$	Effective thermal neutron attenuation length for medium	
	$i [g cm^{-2}]$	
	-1	
	$\Lambda_{th,ss} = \sum_{th=1}^{1} $ (E.33)	
	th,ss	
$L_{th,ss}$	Diffusion length for thermal neutrons in the subsurface	
	$[g \text{ cm}^{-2}]$	
	$L_{th,ss} = \sqrt{\frac{D_{th,ss}}{\Sigma_{th,ss}}} \tag{E.34}$	
	$\bigvee \Sigma_{th,ss}$	
$P_{th,ss,m}$	production rate of nuclide m by thermal neutrons [atoms	
	$[g^{-1}yr^{-1})]$	
$L_{th,a}$	Diffusion length for thermal neutrons in the air $[g \text{ cm}^{-2}]$	
	$D_{th,a}$	
	$L_{th,a} = \sqrt{\frac{D_{th,a}}{\Sigma_{th,a}}} \tag{E.35}$	
	V [⊥] th,a	
$\Phi_{th,ss}(Z)$	Thermal neutron flux at depth Z [neutrons $cm^{-2}yr^{-1}$)]	
	Continued on next page	

Table E.8 – continued from previous page

Term	Definition
$\Phi_{th,ss}^*$	Thermal neutron flux that would be observed at the land
,	surface if the properties of the atmosphere and subsurface
	were identical [neutrons $\rm cm^{-2} \ yr^{-1}$]
	$\Phi_{th,ss}^* = \frac{p(E_{th})_a R_{th,ss} \Phi_{eth,ss}^*}{\Lambda_{eth,ss} (\Sigma_{th,ss} - \frac{D_{th,ss}}{\Lambda_{eth,ss}^2})} $ (E.36)
$\Phi^*_{th,a}$	Thermal neutron flux that would be observed at the land
± th,a	surface if the properties of the medium did not change
	and were identical to the atmosphere [neutrons cm^{-2}]
	yr^{-1}
	$(\Pi) D I^*$
	$\Phi_{th,a}^* = \frac{p(E_{th})_a R_{th,a} \Phi_{eth,a}^*}{\Lambda_{eth,a} (\Sigma_{th,a} - \frac{D_{th,a}}{\Lambda^2})} $ (E.37)
	$\Lambda_{eth,a}(\Sigma_{th,a}-rac{\Lambda_{eth,a}^2}{\Lambda_{eth,a}^2})$
Bu	Ratio of thermal neutron production in the rock to that
$R_{th,ss}$	in the atmosphere [unitless]
	$R_{th,ss} = \frac{p(E_{th,ss})}{p(E_{th,a})} \tag{E.38}$
	Elemental thermal neutron cross-section for the subsur-
$\sigma_{th,ss,k}$	face [barns] 1 barn = $10^{-24} cm^2$
$\sigma_{th,a,k}$	Elemental thermal neutron cross-section for the air
	[barns] 1 barn = $10^{-24} cm^2$
$\Sigma_{th,ss}$	$\begin{array}{c} \text{[barns] f barn = 10} & \text{cm} \\ \text{Macroscopic neutron absorption cross section } [\text{cm}^2 \text{ g}^{-1}] \end{array}$
	$\Sigma_{th.ss} = \sum \sigma_{th.k} N_{k.ss,bulk} = \Lambda_{th.ss}^{-1} \tag{E.39}$
	$\Sigma_{th,ss} = \sum_{k} \sigma_{th,k} N_{k,ss,bulk} = \Lambda_{th,ss}^{-1} $ (E.39)
$\Gamma_{th,ss,m}$	Total rate of thermal neutron absorption [neutrons
	$[g^{-1}yr^{-1})]$
Muons	
	Continued on next page

Table E.8 – continued from previous page

Term	Definition	
f_i^*	Probability for particle emission to the radionuclide. See Heisinger et al. (2002a).	
$f_{i,D}$	Fraction of muons stopped by element k and absorbed by	
J i,D	the nucleus before decay of the muon (Fabryka-Martin,	
C	1988) [unitless]	
$f_{i,C}$	Chemical compound factor [<i>unitless</i>] Chemical com-	
	pound factor (for Be, Al, C, see Heisinger et al. (2002a));	
	values computed on a sample by sample basis for 36 Cl	
	due to variations in composition using values from von	
	Egidy & Hartmann (1982).	
	$f = M_{k,bulk}\Omega_k$ (F 40)	
	$f_{i,C} = \frac{M_{k,bulk}\Omega_k}{\sum j M_{j,bulk}\Omega_j} \tag{E.40}$	
N	A top number density of the target stop [stops π^{-1}]	
Natoms	Atom number density of the target atom [atoms g^{-1}]	
Ω	Atomic number of the element. Subscript k refers to the	
	target element and j refers to all elements in the rock.	
Р	Average probability of muon capture by a nucleus rela-	
	tive to that of oxygen (von Egidy & Hartmann, 1982)	
	[unitless]	
$P_{\mu-,m}$	Production rate for negative muon production of nuclide	
<i>p</i> ² ,	m. See eqns B.3 and B.32.	
$P_{\mu,fast,m}$	Production rate for fast muon production of nuclide m.	
$\mu, j ust, m$	See eqn B.24.	
$P_{n,\mu}$	Production rate for muon-induced neutrons. See eqn	
$\prod n, \mu$	B.29.	
D (7)		
$P_{n,\mu}(Z)$	Total muon-induced neutron production at depth Z [neu-	
	trons cm ⁻² yr ⁻¹]; value at surface is $P_{n,\mu}(0)$	
	$P_{n,\mu}(Z) = Y_s \Psi_{\mu}(Z) + 5.8 \times 10^{-6} \Phi_{\mu f}(Z) $ (E.41)	
	Continued on next page	

Table E.8 – continued from previous page

	Table E.8 – continued from previous page
Term	Definition
$\Phi_{\nu,0}$	Vertical muon flux at SLHL as a function of depth. Only
	valid for depths of $<200,000$ g/cm ² . See equation B.17.
	$[cm^{-2}s^{-1}sr^{-1}]$
$\phi_{\mu f}(Z,\theta)$	Fast muon flux at depth Z [muons $g^{-1}yr^{-1}$]; calculated
	from the muon code
$\Psi_{\mu-}(Z)$	Slow negative muon stopping rate at depth Z [muons g^{-1}
	yr^{-1} ; calculated from the muon code
$R_{\mu-}(h)$	Rate of negative muons stopping at an atmospheric depth
	of h
$R_{\mu}(Z)$	Ratio of muon production to epithermal neutron produc-
- °µ (-)	tion [unitless]
	[
	$B_{el,\mu}P_{n,\mu}(Z) \qquad (E_{el,\mu}P_{n,\mu}(Z))$
	$R_{\mu}(Z) = \frac{S_{el,\mu}P_{n,\mu}(Z)}{S_{el}P_f(0)R_{eth}} $ (E.42)
R'_{μ}	Ratio of the muon production rate to the production rate
$\Gamma \iota_{\mu}$	for thermal neutrons. [<i>unitless</i>] See eqn B.33.
	for thermal neutrons. [<i>unitiess</i>] see equ D.55.
	$p(E_{th})_{a} = (\overline{E}_{th})_{a}$
	$R'_{\mu} = \frac{p(E_{th})_a}{p(E_{th})} R_{\mu} $ (E.43)
	Cross-section for fast muon production at 190 GeV $[mb]$.
σ_{190}	Note: 1 barn= 1×10^{-24} cm ²
Y_s	Average neutron yield per stopped negative muon [neu-
	trons/(stopped negative muon)]
	$V = \sum f \qquad f V \qquad (E 44)$
	$Y_s = \sum_{k} f_{c,k,bulk} f_{d,k} Y_{n,k} \tag{E.44}$
	k
	Average neutron yield per captured muon for element k
$Y_{n,k}$	
Dadiagonia D	- (Fabryka-Martin, 1988)
Radiogenic P	
$F_{k,bulk}$	Fractional abundance of element k in ppm in the bulk
	rock
	Continued on next page

Table E.8 – continued from previous page

P_r Total radiogenic production from all mechanisms is particular sample. (equation B.37) $P_{n,\alpha}$ Production rate of neutrons from alpha particles in n trons/g/yr $P_{n,sf}$ Neutron production rate due to the spontaneous fiss of 238 U S_k Mass stopping power of element k for α -particles of given energyXNeutron production factors related to the light isote composition of the rock matrix. See equation B.40.YNeutron production factors related to the light isote composition of the rock matrix. See equation B.41. Y_n^U Neutron yields of element i per ppm U in equilibrium	
$P_{n,\alpha}$ Production rate of neutrons from alpha particles in n trons/g/yr $P_{n,sf}$ Neutron production rate due to the spontaneous fiss of 238 U S_k Mass stopping power of element k for α -particles of given energyXNeutron production factors related to the light isoto composition of the rock matrix. See equation B.40.YNeutron production factors related to the light isoto composition of the rock matrix. See equation B.41.	eu-
trons/g/yr $P_{n,sf}$ Neutron production rate due to the spontaneous fiss of 238 U S_k Mass stopping power of element k for α -particles of given energyXNeutron production factors related to the light isote composition of the rock matrix. See equation B.40.YNeutron production factors related to the light isote composition of the rock matrix. See equation B.41.	eu-
$P_{n,sf}$ Neutron production rate due to the spontaneous fiss of 238 U S_k Mass stopping power of element k for α -particles of given energyXNeutron production factors related to the light isoto composition of the rock matrix. See equation B.40.YNeutron production factors related to the light isoto composition of the rock matrix. See equation B.41.	
of 238 U S_k Mass stopping power of element k for α -particles of given energyXNeutron production factors related to the light isote composition of the rock matrix. See equation B.40.YNeutron production factors related to the light isote composition of the rock matrix. See equation B.41.	
Sk Mass stopping power of element k for α-particles of given energy X Neutron production factors related to the light isote composition of the rock matrix. See equation B.40. Y Neutron production factors related to the light isote composition of the rock matrix. See equation B.41.	on
given energyXNeutron production factors related to the light isote composition of the rock matrix. See equation B.40.YNeutron production factors related to the light isote composition of the rock matrix. See equation B.41.	fa
composition of the rock matrix. See equation B.40.YNeutron production factors related to the light isotecomposition of the rock matrix. See equation B.41.	
Y Neutron production factors related to the light isotropy composition of the rock matrix. See equation B.41.	pe
composition of the rock matrix. See equation B.41.	
	pe
Y_n^U Neutron yields of element i per ppm U in equilibrium	
Y_n^U Neutron yields of element i per ppm U in equilibrium Y_n^{Th} Neutron yields of element i per ppm Th in equilibriu	n
Accumulation	
D Depth of the sample with 'old' representing the original depth of the sample with 'old' representing th	nal
sample depth and 'new' is the updated sample dep	th,
accounting for erosion during the time period	
Δt Time step in the CRONUScalc program.	
ϵ Erosion rate [g/cm ²]	
$\int f_{decay} \qquad \qquad \text{Decay factor that accounts for the fact that some of}$	he
nuclides produced at the beginning of the time per	od
will have decayed by the end of the period. (equation	8)
λ Decay constant for the nuclide	
N_{tot} Total inventory in the sample up to the current time s	ep
N_{prev} Inventory from all previous time steps.	
P_{tot} Instantaneous production rate of the nuclide from	
mechanisms and is the sum of production from all ot	
mechanisms.	ner

Table E.8 – continued from previous page

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