CRONUS-Earth cosmogenic ^{36}Cl calibration

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Abstract

Chlorine-36 production rates obtained from different geological calibration studies (e.g. Evans et al., 1997; Phillips et al., 2001; Schimmelpfennig et al., 2011; Stone et al., 1996; Swanson and Caffee, 2001) vary significantly, principally because of the many reactions contributing to the production of this nuclide. The CRONUS-Earth Project has provided high-quality geological calibration sites, including Lake Bonneville, Peru, and Scotland, for a large-scale calibration of ^{36}Cl production rates. Three sites were used to calibrate the K and Ca spallation pathways for ^{36}Cl production yielding production rates of 56.0 ± 4.1 at ^{36}Cl (g Ca) $^{-1}$ yr $^{-1}$ and 155 ± 11 at ^{36}Cl (g K) $^{-1}$ yr $^{-1}$ respectively, using Lifton-Sato-Dunai scaling (LSDn). The low-energy production parameter, $P_f(0)$, was calibrated separately using CRONUS-Earth data from the Bonneville and Baboon Lakes sites where Cl concentrations were higher, and yielded a value of 759 ± 180 neutrons (g air) $^{-1}$ yr $^{-1}$. There is significant uncertainty associated with this pathway due to the sensitivity of this reaction to environmental conditions. The uncertainties associated with the calibrated production parameters

were estimated based on the variance of calculated ages from independent ages for an independent secondary dataset.

Keywords: cosmogenic nuclides; exposure age; ^{36}Cl ; production rate calibration; low energy neutron absorption

1. Introduction

A premier use of cosmic-ray-produced (cosmogenic) nuclides is the determination of exposure ages and erosion rates, both of which require precise knowledge of production rates. The CRONUS-Earth (Cosmic-Ray prOduced NUclide Systematics on Earth) Project was funded to improve our knowledge of cosmogenic nuclide systematics. A significant outcome of the CRONUS-Earth Project was the calibration of terrestrial cosmogenic nuclide production rates using geological calibration locations. This paper describes the CRONUS-Earth geological calibration effort for ^{36}Cl .

Of the most commonly measured cosmogenic radionuclides - ^{10}Be , ^{26}Al , and ^{36}Cl - the ^{36}Cl production rate is the most difficult to calibrate owing to the many production reactions that must be accounted for - spallation, low-energy neutron absorption, and muon reactions - and the numerous possible target elements; K, Ca, Cl, Ti, and Fe. Table 1 presents a summary of 20 years of calibration studies. There are glaring discrepancies and many mechanisms for these discrepancies have been proposed: problematic independent ages and/or erosion (Schimmelpfennig et al., 2014; Swanson and Caffee, 2001); incorrect scaling models (Swanson and Caffee, 2001); improper partitioning of reaction mechanisms (Alfimov and Ivy-Ochs, 2009; Schimmelpfennig et al., 2009); and sensitivity to hydrogen (Dunai et al., 2014; Licciardi et al., 2008; Swanson and Caffee, 2001).

The calibrated nuclear reactions (and production parameters) considered in this study are: spallation production from Ca ($P_{s,\text{Ca}}$) and K ($P_{s,\text{K}}$); muon production from Ca ($\sigma_{0,\text{Ca}}$ and f_{Ca}^*) and K ($\sigma_{0,\text{K}}$ and f_{K}^*); and production from low-energy neutron absorption by ^{35}Cl ($P_f(0)$). The calibrated production rate parameters are discussed in terms of previous calibrations and are validated against a secondary dataset, with extensive discussion regarding the uncertainties, limitations, and sensitivity of sample ages to various inputs.

Source	Location	Landform	Elevation	Lat	Age	Meth	Prod Rate
Phillips et al. (2001)	Utah	Basalt flow	1445	39N	16.5	WR	Ca: 66.8 ± 6.8 K: 137 ± 60 P _f (0): 626 ± 43
	Idaho	Basalt flow	1367-1798	44N	2.1-13.7		
	New Mexico	Basalt flow	2058-2578	35N	3-18.2		
	Wales	Boulder on beach	375	52N	11.8		
	Ellesmere Is	Erratic boulder	80-100	80N	9		
	NW Territories	Glacial bedrock	20	68N	14.8		
	Arizona	Meteor crater ejecta	1730	35N	49		
Evans et al. (1997)	Sierra Nevada	Glacial bedrock	3000-3600	38N	13.1	MS	K: 170 ± 25 K: 241 ± 9
	Scotland	Glacial bedrock	520	58N	11.6		
	Antarctica	Bedrock	2000-2200	70S	SS		
Stone et al. (1996)	Utah	Basalt flow	1445	39N	17.3	MS	Ca: 48.8 ± 4.8
Evans (2001)	Scotland	Glacial bedrock	520	58N	11.6	MS	P _f (0): 740 ± 63
Swanson and Caffee (2001)	WA State	Moraine boulders and glacial bedrock	10-140	48N	15.0	WR	Ca: *91 ± 5 K: *228 ± 18 P _f (0): 762 ± 28
	Iceland	Basalt flow	30-500	64N	4-10	WR	Ca: 57 ± 5
Alfimov and Ivy-Ochs (2009)	Reanalysis of Liu et al. (1994)	Concrete block	NA	NA	NA	NA	P _f (0): 760 ± 150
Schimmelpfennig et al. (2011)	Sicily Argentina	Basalt flow Basalt flow	2000 2300-2500	38N 36S	10 15	MS	Ca: 42.2 ± 4.8 K: 124.9 ± 8.1
Braucher et al. (2011)	France	Bedrock	310	43N	310	WR	Ca: 42 ± 2.2
Schimmelpfennig et al. (2014)	Swiss Alps	Landslide	820	46N	34	MS	K: 145.5 ± 7.7

Table 1 - Summary of previous ^{36}Cl calibration studies. Elevation is in meters. ‘Meth’ is the sample type, either whole rock (WR) or mineral separate (MS). ‘Prod rate’ is the value produced by the study (as published). All studies here used Lal (1991) or Lal (1991)/Stone (2000) scaling and units are atoms (g Ca) $^{-1}$ yr $^{-1}$ for Ca, atoms (g K) $^{-1}$ yr $^{-1}$ for K, and neutrons (g air) $^{-1}$ yr $^{-1}$ for $P_f(0)$. *Indicates production rate includes muons. SS indicates steady state.

2. Background

2.1. Scaling Models

Scaling models allow site-specific production rates to be extended to other geographic locations or to other times. Numerous scaling models have been proposed to correct for latitude and longitude, elevation, atmospheric pressure anomalies, geomagnetic field changes, dipole and non-dipole effects, and solar modulation. Production rates based on geological calibrations are completely dependent on the scaling model used to produce the results. Previous ^{36}Cl production rate calibrations used the Lal (1991) or Lal (1991)/Stone (2000) (abbreviated St) scaling model; the geomagnetically-corrected version of the St scaling model (Nishiizumi et al., 1989) is referred to as ‘Lm’. Later, scaling models were proposed that were all based on data from the global neutron-monitor network: Du (Dunai, 2000, 2001), Li (Lifton et al., 2008; Lifton et al., 2005), and De (Desilets and Zreda, 2001; Desilets et al., 2006). The new Lifton-Sato-Dunai (LSD) physics-based scaling model (Lifton et al., 2014) developed as part of the CRONUS-Earth Project, provides a flux-based version (LSD) as well as a second version that incorporates nuclide-dependent and energy-dependent reaction cross-sections into the scaling model, producing nuclide-dependent scaling factors (LSDn). In the CRONUS-Earth calibration, the LSD models (LSD and LSDn), along with the Lal-based scaling models, fit the calibration data for nuclides with wide global distribution (e.g., ^{10}Be and ^{26}Al) better than do the neutron monitor-based scaling models (Borchers et al., 2015). We present and interpret results from the nuclide-dependent LSDn model, but include the production rates for all seven scaling models for comparison in Table 3 (spallation production rate and $P_f(0)$) and Table 5 (muon parameters).

Production rates are reported as the modern-day production rate, scaled (using the same method used

for the calibration) to the reference point of sea level and high latitude. (Note that in CRONUScale (described in Section 4.2 with full details in Marrero et al., 2015, this issue) the LSD and LSDn scaling methods are denoted “Sf” and “Sa”, respectively.)

2.2. Production mechanisms

The production of a cosmogenic nuclide is dependent on the sample’s position in space (i.e., dip and direction of dip of the sampled surface) and the incoming cosmic-ray flux, which varies as a function of geographic position and time (Nishiizumi et al., 1989; Gosse and Phillips, 2001). The production also typically decreases with increasing depth in the sample material due to attenuation of the cosmic-ray flux through interactions with matter. These interactions of cosmic rays with matter produce cosmogenic nuclides through three main categories of reactions: spallation, interactions with muons, and low-energy neutron absorption.

2.2.1. Spallation

Spallation reactions caused by high-energy neutrons colliding with the nuclei of atoms in minerals dominate cosmogenic nuclide production at the surface. The spallogenic production of cosmogenic nuclides is given by Equation 1, which reflects the assumption that nuclide production from this pathway decreases exponentially with depth.

$$P_s(Z) = S_T \sum S_{el,s} P_k(0) C_k \exp\left(-\frac{Z}{\Lambda_{f,e}}\right) \quad (1)$$

P_s is the production rate from a spallation reaction in a sample; P_k is the production rate of ^{36}Cl by spallation of element k at sea level and high latitude; Z is the depth (in mass-length units: g cm^{-2}); S_T is the topographic shielding factor; $S_{el,s}$ is the elevation-latitude-longitude scaling factor for the particular reaction of interest (“s” for spallation in this case); C_k is the concentration of the target element k ; and $\Lambda_{f,e}$ is the effective attenuation length for fast neutrons. The production is summed for all target elements k that produce ^{36}Cl to give the total spallation production rate.

Chlorine-36 is produced from Ca, K, Fe, and Ti. For most common rock types the combined contributions from Ti and Fe are typically less than 1-2% of the total ^{36}Cl production, but for oxide mineral separations it could be much larger. The samples used in this calibration study have low Fe

and Ti, so accordingly the calibration focused on Ca and K. The assigned spallation production rate of Fe is 1.9 at $^{36}\text{Cl} \text{ (g Fe)}^{-1} \text{ yr}^{-1}$ (Stone et al., 2005). The nominal value for the production from Ti, 3.8 at $^{36}\text{Cl} \text{ (g Ti)}^{-1} \text{ yr}^{-1}$, was derived by scaling the Fe production rate of Stone (2005) by the relative Ti/Fe production ratio predicted by Masarik (2002). Fink et al. (2000) obtained a value of 13 ± 3 at $^{36}\text{Cl} \text{ (g Ti)}^{-1} \text{ yr}^{-1}$, higher than the value used in this study. Our samples have little Ti so the effective difference between these two is insignificant on our calibration.

2.2.2. Muons

Although production of cosmogenic nuclides by muons is small at the surface compared to spallation production, it can be significant for samples taken from depth profiles, for samples collected in areas having high erosion, or in samples in which inheritance is significant (Stone et al., 1998). Muons penetrate very deeply into the subsurface, as shown by their very large apparent attenuation length ($\sim 1300 \text{ g cm}^{-2}$, compared to $\sim 150 \text{ g cm}^{-2}$ for spallation) (Braucher et al., 2011; Heisinger et al., 2002b). In the CRONUScalc program, muon production is calculated using the production equations from Heisinger et al. (2002a, b) scaled according to Lifton et al. (2014). The production rate from fast muons is shown in Equation 2.

$$P_{\mu,fast} = \phi_{\mu,total} \beta(Z) (\bar{E}(Z))^\alpha \sigma_0 N_{t,i} \quad (2)$$

$P_{\mu,fast}$ is the fast-muon production rate; β and $\bar{E}$ are depth-dependent constants described in Heisinger et al. (2002b); $N_{t,i}$ is the concentration of the target element in atoms g^{-1} ; $\phi_{\mu,total}$ is the total muon flux in units of muons $\text{cm}^{-2} \text{ yr}^{-1}$; and α is an empirically fitted exponent. Heisinger et al. (2002b) used a value of 0.75 for the exponent α based on AMS experiments, but experimental results are permissive of a range of values of α between 0.75 and >1.0 (Heisinger et al., 2002b); the CRONUS-Earth project has chosen a value of $\alpha = 1.0$. This decision is supported by the value of α near 1 calibrated by Braucher et al. (2011) from a ^{10}Be profile. The parameter, σ_0 , is the nuclide production cross-section in 10^{-24} cm^2 at 1 GeV and is the fitted parameter for fast muon production in the model. As a calibration starting point, the laboratory-measured value of σ_{190} (related to σ_0 through the equation $\sigma(E) = \sigma_0 E^\alpha$,

where E is the muon energy, e.g. 190 GeV) is presented in Heisinger et al. (2002b) for Ca; this value was not given for K.

Slow muon production is calculated according to Equation 3 (Heisinger et al., 2002a). In this equation, $\phi_{\mu-}$ is the negative muon stopping rate in units of stopped muons $\text{g}^{-1} \text{yr}^{-1}$, which is calculated using the model by Lifton et al. (2014); $f_{i,C}$ is the compositionally dependent compound factor; and $f_{i,D}$ is the probability that the negative muon does not decay in the K-shell before it is captured by the nucleus. The parameter f_i^* , the calibrated slow muon production rate parameter, is the effective probability for particle emission to the nuclide of interest after the negative muon is captured. Values of f_i^* based on laboratory experiments are provided in Heisinger et al. (2002a) for ^{36}Cl (both for K and for Ca).

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

Muons can produce ^{36}Cl through interactions with Ca, K, Fe, and Ti. For the main two targets, Ca and K, previously published depth profiles allow the reparameterization of the muon models for slow and fast muon reactions. There are currently no studies or models to predict the production of ^{36}Cl by muons from Fe and Ti. These production rates are assumed to be only a small fraction of those for spallation so they are considered insignificant and are not included in the production model at this time. Parameterization of these reactions will require studies that measure ^{36}Cl in magnetite or garnets in deep or highly eroded rock.

2.2.3. Low-energy neutron absorption

Thermal and epithermal neutrons (grouped here as low-energy neutrons) can be absorbed by ^{35}Cl to produce ^{36}Cl . This pathway is complicated by the sensitivity of the magnitude of the low-energy neutron flux to the concentration of hydrogen in the environment (most commonly present as pore water in the rock or soil, but also as snow and ice cover immediately above the rock or soil surface, and occasionally in hydrated minerals) and the chemical composition of the sample. These reactions are discussed in detail in both Marrero et al. (2015, this issue) and Gosse and Phillips

(2001). Equation 4 shows the simplified production equation. This equation is generalized in order to apply to either thermal or epithermal neutrons.

$$P_e(Z) = \frac{f_e}{\Lambda_{e,ss}} \Phi_{e,ss,total} \quad (4)$$

P_e is the production rate in a sample for a specific pathway; e indicates the energy level of the neutron flux (either thermal or epithermal); $\Phi_{e,ss,total}$ is the epithermal or thermal neutron flux in the subsurface; f_e is the fraction of absorbed neutrons of energy e that are taken up by the target element to produce ^{36}Cl ; and $\Lambda_{e,ss}$ is the effective attenuation length for neutrons of energy e . The neutron-flux term is calculated from equations that account for the diffusion of low-energy neutrons from the rock to the atmosphere at the interface between these (Gosse and Phillips, 2001; Marrero et al., 2015, this issue). These low-energy neutrons originate principally from the moderation of high-energy cosmic-ray neutrons, but also to a minor extent from neutrons produced by muon reactions.

The parameter calibrated for the low-energy reactions is denoted $P_f(0)$. This parameter, defined as the epithermal neutron production rate in the atmosphere directly above the atmosphere/subsurface interface, with units of neutrons $(\text{g air})^{-1} \text{ yr}^{-1}$, is what ultimately quantifies the low-energy neutron flux. As the fast neutron flux is moderated by nuclear interactions with the sample, the fast neutrons are moderated into the thermal and epithermal neutron fluxes, whose magnitudes also depend on the composition of the rock. $P_f(0)$ is chosen as the calibration parameter specifically because, being defined in the atmosphere (which is assumed to be chemically invariant), it is not compositionally dependent.

Chlorine-36 is also produced by an intermediate-energy neutron reaction (1 to 10 MeV) off of K (Johnson et al., 1967; Reedy, 2013). This is explicitly accounted for in the CRONUScalc energy-dependent production model (LSDn; Lifton et al., 2014, as implemented by Marrero et al. 2015, this issue) and is implicitly included in the spallation-production term for K in the other production (i.e., scaling) models.

3. Calibration Sites

The CRONUS-Earth sampling sites are classified as either ‘primary’ or ‘secondary’ calibration sites. Primary sites have robust independent age control, continuous exposure to cosmic rays, simple geometry, and minimal or no erosion. Sites that did not meet the strict primary site guidelines were labelled ‘secondary’ sites. Additional high-quality sites from the literature, mostly previous calibration efforts, were included in the secondary dataset as well. These secondary calibration datasets were updated as necessary; in some cases the original authors were contacted to verify the information or provide missing details for the samples. Only studies produced prior to 2011 were included in the secondary dataset as this was the date of the conclusion of data acquisition of the CRONUS-Earth Project and the initiation of the statistical analyses. Some studies after that date are included in the comparison table, but are not in the secondary dataset. The secondary calibration dataset is completely independent of the primary calibration dataset so it can be used to assess the fit of the calibrations, as well as empirically estimate the uncertainty assigned to the calibrated production parameters. Some sites were excluded from the secondary dataset owing to extremely large age uncertainties (Schimmelpfennig et al., 2009) or concerns over applicability to other locations (Licciardi et al., 2008). For complete details of the site categorization and results from the excluded sites, see Marrero (2012).

Each site used in the main calibration is summarized here while the secondary site descriptions can be found in Appendix 2. In a few cases, some samples were excluded from the calibration. A table summarizing these choices is also in Appendix 2, with a complete spreadsheet available in the supplemental material. The spreadsheet includes all relevant sample information for all samples used in this paper, including location information, chemical composition, analytical uncertainties, and other calculator inputs required for CRONUScalc (Marrero et al., 2015, this issue). The processing laboratory is indicated for samples using the following abbreviations: University of Washington (UW), New Mexico Tech (NMT), Dalhousie University (DAL), and Purdue PRIME Lab (PRIME). All calibrated radiocarbon ages presented here are presented as years before 2010 and were calculated using the Calib 6.0 radiocarbon calibration program (Stuiver and

Reimer, 1993; Stuiver et al., 2005) with the IntCal 09 curve option and one-sigma uncertainties; no marine reservoir corrections were required as all of the radiocarbon dated media were terrestrial.

3.1. Tabernacle Hill, Utah (CRONUS primary site)

The Tabernacle Hill basalt flow (39°N and 1450 m elevation) erupted into the Provo level of Pleistocene Lake Bonneville (Gilbert, 1890). The time of this eruption is tightly bracketed by maximum limiting radiocarbon ages on wood and charcoal (Marrero, 2012; Oviatt et al., 1992) for the Bonneville flood (as determined by a Bayesian analysis of 19 radiocarbon ages) and by minimum limiting ages of tufa collected from the edges of the basalt flow (Lifton et al., 2015; Oviatt et al., 1992). The limiting ages constrain the eruption of the basalt to $18,200 \pm 300$ years (for measurement and calibration details refer to Lifton et al., 2015). A single erosion rate of 1 ± 1 mm kyr⁻¹ was assigned to the sampled basalt bedrock surfaces at this site based on well-preserved pahoehoe ropes, with a large uncertainty assigned based on observed local variability. Ten Ca-rich plagioclase mineral separates were prepared by a single lab (UW) from this site as well as 24 whole-rock samples that were prepared by three different labs (DAL, NMT, PRIME).

3.2. Huancané, Peru (CRONUS primary site)

The Huancané, Peru, site is located in the central Peruvian Andes at approximately 13°S and 4850 m elevation. Advances of the Quelccaya ice cap during the late Pleistocene left several sets of terminal and recessional moraines. The CRONUS Earth samples were collected from boulders from the Huancané II moraines of Mercer and Palacios (1977). The age of these moraines is well-constrained by radiocarbon dates from peats below and above the moraines, and by basal ages from lake cores on either side of them (Kelly et al., 2015; Kelly et al., 2012; Phillips et al., 2015, this issue-c). The maximum and minimum bracketing ages based on ~30 new radiocarbon ages yielded a site age of $12,320 \pm 110$ years before 2010 (calibration and statistical evaluation of the radiocarbon ages has been described by Kelly et al., 2012). Erosion rates were assigned based on a combination of measurements of erosion-resistant protrusions, whose height above the boulder surface was measured on each individual sample boulder, and the results for minimum erosion rates calculated from comparisons of ¹⁰Be concentrations in eroded and glacially polished sections from boulders at the site

(Kelly et al., 2015). Despite the use of two methods to constrain erosion rates at this site, the resulting values were not negligible (3.2-8.1 mm kyr⁻¹, median of 4.8 mm kyr⁻¹; see supplemental spreadsheet) and we recognize that there may still be a small amount of uncertainty introduced from this parameter. For comparison, production rates for the potassium pathway determined with zero erosion for all samples using LSDn scaling are 4% lower than the result calculated using the erosion rates assigned here. However, the obvious weathering on the boulders makes the zero erosion scenario unrealistic, indicating that the uncertainty contributed from the assigned erosion rate remains relatively small. The cosmogenic ³⁶Cl samples prepared at NMT produced anomalously low concentrations and high scatter (see Marrero, 2012) so the Huancané whole rock and mineral separate samples from NMT were not used in the final calibration for ³⁶Cl. The site description is provided in Phillips et al. (2015, this issue-c). From this site, there were ten K-rich feldspar mineral separates prepared by a single lab (UW).

3.3. Scotland (CRONUS primary site)

Sites on the Isle of Skye and Scottish Highlands constitute the primary Scotland site. The Isle of Skye, Scotland site is located in the Inner Hebrides along the northwest coast of Scotland. Additional samples were collected on the mainland, in the northwestern Scottish Highlands. The samples used in this calibration are from Coire Fearchair, Beinn na Caillich, Scotland located at ~57°N with sample elevations ranging from 300 to 450 m. These samples were collected from rockfall boulders composed of the local lithology, hornblende granite. The boulders were carried either sub- or supra-glacially away from the failure wall, where most Scottish rockfall deposits accumulate (Ballantyne and Stone, 2004), but had not yet reached the terminal moraines, indicating that they were initially exposed upon deglaciation at the end of the Loch Lomond readvance (Ballantyne, 2008; Sissons, 1979). There is a significant amount of information constraining the age of the beginning and end of the Loch Lomond Stade, including sediment analysis in lake cores (Brooks et al., 2012; Walker et al., 1988), pollen analysis (e.g. Vasari, 1977; Walker and Lowe, 1990; Williams, 1977), temperature reconstructions from chironomids (Brooks et al., 2012), varve comparisons (MacLeod et al., 2011; Palmer et al., 2012), and radiocarbon dates on plant macrofossils (MacLeod et al., 2011) and other materials (e.g. McIntyre et al., 2011). This information was compared to the more geographically

distant Greenland Ice Core (Rasmussen et al., 2006) and models of the local glaciation in Scotland (Golledge et al., 2008). A local reconstruction of the Loch Lomond Stadial glaciation and deglaciation was synthesized, primarily relying on the most local information available (Phillips, 2015, pers. comm.). The exposure age assigned to these samples is $11,700 \pm 300$ years based on the synthesized chronology. Many samples collected in Scotland as part of the project still contained evidence of glacial erosion (straie) suggesting negligible post-glacial erosion, but small erosion rates were applied to samples 06SKY06, -07, -08. Only four K-rich feldspars, all prepared by a single lab (UW), were included from this site.

3.4. Baboon Lakes (CRONUS secondary site)

Although technically classified as a secondary site, this was the only CRONUS-sampled site that had production dominated by low-energy neutron absorption by Cl and some samples from this site were used in the calibration of $P_f(0)$. The Baboon Lakes are located in the Sierra Nevada, CA, USA at a latitude of 37°N and an elevation of 3400 m. The features that were sampled are boulders on moraines interpreted to represent the Recess Peak advance (Birman, 1964; Clark and Gillespie, 1997). The chronology presented in Phillips (2015, this issue) is based primarily on radiocarbon ages of organic matter from lake cores within the glacial limits, but not directly at the sample site. The site was assigned an age of 13310 ± 250 years before 2010. The sample sites are described in Phillips et al. (2015, this issue-b). This site is not ideal due to heavy winter snow accumulation. Small snow corrections were performed for all three samples but contribute little additional uncertainty (see Section 6.3). The rounded boulders were assigned an erosion rate of $1 \pm 1 \text{ mm kyr}^{-1}$, with the large uncertainty owing to the possibility of negligible erosion since exposure. Three Cl-rich hornblende mineral separates, all prepared by a single lab (NMT), were used from this site.

3.5. Sample composition summary

Although the CRONUS-Earth Project produced data for numerous geological calibration locations, the existence of three major production reactions for ^{36}Cl means that it requires more calibration samples than any other nuclide in order to constrain all the production rates. Based on the minerals available for separation and analysis at each site, there was only one site to constrain the

$^{36}\text{Cl}_{(\text{Ca})}$ pathway (Tabernacle Hill) and two sites each to constrain the $^{36}\text{Cl}_{(\text{K})}$ (Peru and Scotland) and $^{36}\text{Cl}_{(35\text{Cl})}$ (Baboon Lakes and Tabernacle Hill) pathways. The relative contributions of each production pathway for all the primary and secondary sites are graphed in Figure 1. Table 2 contains all the ^{36}Cl -producing elements for the samples used in the main calibration.

{Figure 1: This figure is to be black and white in printed copy and color online. 1-column width. Color and black & white versions of this figure are provided in separate files at the end. Contact author if higher-quality image is required.}

Figure 1: Ternary diagram showing the the relative contribution from each main production pathway for the CRONUS-Earth samples. Muons are included for production from Ca and K pathways. The figure includes CRONUS-Earth sites used in the calibration dataset as well as those included in the secondary dataset. Non-CRONUS sites included in the secondary dataset are shown as unlabeled smaller blue circles. Abbreviations refer to the site: TAB - Tabernacle Hill, UT; BL - Baboon Lakes, CA; Peru - Huancané, Peru; SCOT - Isle of Skye, Scotland; New England - Littleton-Bethlehem moraine. Additional abbreviations refer to the composition of the samples: MS - plagioclase or K-feldspar mineral separates; WR - whole rock. Site descriptions are included in Sections 3.1-3.4 and Appendix 2, with composition available in Appendix 1 and the supplemental material.

	Name -	K ₂ O wt %	CaO wt %	TiO ₂ wt %	Fe ₂ O ₃ wt %	Cl ppm
Tabernacle Hill (MS)	TH-1f	0.24	12.98	0.08	0.62	2.9 ± 0.9
	TH-2f	0.23	13.28	0.08	0.6	2.2 ± 0.8
	TH-2c	0.23	13.28	0.08	0.6	5.0 ± 1.0
	TH-3f	0.23	13.48	0.06	0.6	3.9 ± 1.1
	TH-3c	0.23	13.48	0.06	0.6	2.8 ± 0.9
	05-TAB-01-PLAG-NXHN	0.25	13.06	0.08	0.62	11.6 ± 1.3
	05-TAB-03-PLAG-NXHN	0.23	13.23	0.08	0.63	15.4 ± 2.4
	05-TAB-04-PLAG-NXHN	0.24	13.1	0.08	0.62	10.9 ± 1.2
	05-TAB-05-PLAG-NXHN	0.23	13.07	0.07	0.61	3.0 ± 1.2
	05-TAB-06-PLAG-NXHN	0.24	13.15	0.08	0.61	9.4 ± 5.1
Peru (MS)	HU08-01-SAN	10.22	0.19	0.02	0.1	0.7 ± 4.5
	HU08-02-SAN	10.25	0.23	0.02	0.03	0.5 ± 4.0
	HU08-03-SAN	10.04	0.33	0.04	0.04	0.0 ± 3.0
	HU08-04-SAN	10.34	0.19	0.02	0.02	0.0 ± 3.7
	HU08-06-SAN	9.92	0.23	0.04	0.04	0.7 ± 3.2
	HU08-10-SAN	9.84	0.25	0.02	0.02	0.6 ± 2.1
	HU08-11-SAN	9.24	0.28	0.03	0.03	0.6 ± 2.6
	HU08-14-SAN	9.52	0.25	0.02	0.02	2.2 ± 3.7
	HU08-15-SAN	9.85	0.25	0.04	0.04	1.9 ± 4.0
	HU08-16-SAN	9.55	0.28	0.06	0.05	2.4 ± 4.5
Scotland (MS)	06-SKY-01-KFSP-NXHN	5.79	0.22	0.02	0.08	38.7 ± 2.1
	06-SKY-06-KFSP-NXHN	5.91	0.14	0.02	0.05	7.4 ± 3.0
	06-SKY-07-KFSP-NXHN	6.28	0.2	0.02	0.12	10.7 ± 2.2
	06-SKY-08-KFSP-NXHN	7.09	0.16	0.01	0.05	22.9 ± 2.3
TAB (WR) - NMT	05TAB01-1	0.75	9.6	1.63	11.02	108.6 ± 1.8
	05TAB01-3	0.76	9.66	1.64	10.94	96.2 ± 1.6
	05TAB05-1	0.68	9.61	1.66	11.11	86.6 ± 1.2
	05TAB05-2	0.68	9.54	1.63	11.14	88.6 ± 1.4
	05TAB05-3	0.67	9.54	1.65	11.21	82.4 ± 4.6
	05TAB06-4	0.75	9.57	1.67	11.28	103.5 ± 1.5
	05TAB06-5	0.74	9.49	1.65	11.3	120.8 ± 1.7

	05TAB06-6	0.74	9.65	1.65	11.14	120.4 ± 1.9
	05TAB07-1	0.87	9.67	1.68	10.91	125.0 ± 2.2
	05TAB07-2	0.87	9.65	1.67	11.03	128.2 ± 2.2
	05TAB07-3	0.87	9.68	1.69	10.97	115.5 ± 1.9
TAB (WR) -DAL	05 TAB 01 bag14	0.78	9.34	1.6	11.74	76.4 ± 1.5
	05 TAB 02 bag18	0.85	10.07	1.72	9.89	93.4 ± 2.9
	05 TAB 03 bag 13	0.86	9.2	1.53	11.63	108.6 ± 1.8
	05 TAB 04 bag14	0.69	9.1	1.51	11.45	74.5 ± 1.4
	05 TAB 05bag16	0.7	9.35	1.59	11.13	68.8 ± 1.4
	05 TAB 06 bag16	0.8	9.38	1.61	11.69	87.4 ± 1.4
	05 TAB 07 bag19	0.89	9.53	1.63	11.41	106.3 ± 1.8
TAB (WR)- PRIME	05TAB01WR	0.78	9.49	1.50	10.61	77 ± 1
	05TAB03WR	0.83	9.49	1.51	10.90	96 ± 2
	05TAB04WR	0.69	9.18	1.55	11.82	76 ± 1
	05TAB05WR	0.71	9.33	1.61	11.32	65 ± 1
	05TAB06WR	0.74	9.35	1.62	11.56	78 ± 5
	05TAB07-1	0.87	9.39	1.62	11.68	103 ± 2
Baboon Lakes (MS)	BBL07-02	2.79	7.84	2.03	26.27	687 ± 40
	BBL07-03	3.04	7.09	2.68	24.74	339 ± 21
	BBL07-01	0.71	10.55	1.91	26.19	533 ± 22

Table 2-Compositions of samples used in the main calibration. These are target compositions, not the bulk rock, and only represent the portion of the rock that was actually dissolved and measured. Samples are labeled as either mineral separate (MS) or whole-rock (WR) samples. Note that the Baboon Lakes (MS) samples are high-Cl mineral separates. For additional calibration sample composition information, see Appendix 1 and Lifton et al. (2015). Samples used in the secondary dataset are included in the spreadsheet in the supplementary material.

4. Methods

4.1. Laboratory Methods

The samples used in this study were processed by four different labs and then measured at two different Accelerator Mass Spectrometry (AMS) facilities. The four processing labs were University of Washington (UW), New Mexico Tech (NMT), Dalhousie University (DAL), and PRIME Lab (PRIME). In general, the procedures follow the same steps for sample preparation, but with slightly varying lab-specific details. After any necessary mineral separation (typically performed using heavy liquids), the silicate samples are leached in dilute nitric acid for a period of time and then rinsed and dried. The samples are then weighed, and

carrier is added to the sample. Most labs use ^{35}Cl -enriched carrier, but UW uses ^{37}Cl -enriched carrier and performs the Cl determination on a separate sample split. The samples are dissolved using a combination of HF and HNO_3 acids, then purified by sulfur removal using BaNO_3 , although the length and number of repetitions of this step varies greatly between labs. Finally, AgCl is reprecipitated, rinsed, and dried. The AgCl was sent for AMS analysis at either PRIME Lab (PRIME, NMT, and DAL samples) or at the Australian National University (UW samples).

The procedures used by New Mexico Tech are given in Marrero (2012). University of Washington procedures, originally presented in Stone et al. (1996), are available on the lab website (Stone, 2001). Dalhousie also uses procedures based on those from the University of Washington (Evans et al., 1997; Stone et al., 1996). The PRIME Lab uses procedures based on Sharma et al. (2000).

Each lab reported blank-subtracted values for ^{36}Cl and Cl concentrations as well as other necessary information. Blank subtractions were generally less than 3%. The individual labs were also responsible for the determination and reporting of all the target element (final, dissolved composition of the sample) and bulk rock (original rock, prior to any mineral separation) compositional information. The composition was determined using a combination of XRF (X-Ray Fluorescence), ICP (Inductively-Coupled Plasma), and NAA (Neutron Activation Analysis). Owing to analyses by different labs, there are sometimes small differences in the sample compositions. Site-specific parameters, such as location, and certain sample parameters, like thickness, were applied consistently to all the samples when multiple labs analysed the same sample. Attenuation lengths have been updated from the values provided in Lifton et al. (2015) and Phillips et al. (2015, this issue-c) and now uses the values found using the table available in Marrero et al. (2015, this issue), using the cutoff rigidity

calculated over the exposure age of the sample with small adjustments for the shielding of the sample.

Although no official interlaboratory comparison for ^{36}Cl has been completed as part of the CRONUS-Earth project at this time, whole-rock field samples processed by three labs were statistically analyzed to look at the intra- and inter-laboratory variability of the results. There was some variability in the whole-rock ^{36}Cl concentrations reported by the labs, with results from different labs varying by up to 25-30% (Lifton et al., 2015), although a portion of this variability is likely due to small variations in the chemical composition of the measured sample. Further analysis of the concentrations by Phillips et al. (2015, this issue-a) indicates that, as a group, the Tabernacle Hill samples were homogenous and within-laboratory random dispersion is the main source of the spread in the dataset.

4.2. Calibration Method

All the calibrations were performed using CRONUScalc (Marrero et al., 2015, this issue). The MATLAB-based code is designed to be versatile, allowing for calibrations, exposure age calculations for surface and depth samples, and erosion rate calculations as well as simple scaling and production rate calculations. The program applies identical code, such as scaling and integration methods, to multiple nuclides (^{26}Al , ^{10}Be , ^{14}C , ^3He , ^{36}Cl , with ^{21}Ne in testing). CRONUScalc was created by the CRONUS-Earth project for use in calibrating production rates and is currently the only available program that incorporates multiple scaling models, including Lifton et al. (2014), and is applicable to ^{36}Cl . The online calculator for surface exposure ages for ^{36}Cl and other nuclides is available at <http://web1.ittc.ku.edu:8888/>, with the complete code repository available for download at <https://bitbucket.org/cronusearth/cronus-calc>.

The ^{36}Cl calibration was performed in a stepwise manner in order to properly calibrate muogenic production and to take advantage of the high-quality mineral separates in the spallation calibration dataset. To reduce the uncertainty from the inclusion of the low-energy production mechanism

(Schimmelpfennig et al., 2009), the spallation production rates were calibrated using a low-chlorine (mineral separate) subset of the primary calibration dataset, as described in Borchers et al. (2015). Ca and K production rates were calculated independently. During this step, the $P_f(0)$ value was fixed at the value determined by Marrero (2012) of 704 neutrons (g air)⁻¹ yr⁻¹. The value of $P_f(0)$ has essentially no effect at this stage due to the very low Cl concentrations in the mineral separates (Cl concentrations typically <10 ppm, see Table 2). The calibration, performed by comparing measured versus predicted ³⁶Cl concentrations, was set up as a nonlinear least squares problem and was solved by the Levenberg-Marquardt method (Aster et al., 2012; Levenberg, 1944; Marquardt, 1963). The goodness-of-fit was assessed using a chi-squared test and interpreted using the resulting p-value (ideally acceptable fits have a p-value >5% (Aster et al., 2012)). Analogous calibration and statistical methods were used for the muon and $P_f(0)$ calibrations as well.

After the spallation calibrations were complete, depth profiles were used to reparameterize the new muon model (described in Section 5.2). The Ca and K muon calibrations were performed sequentially. The spallation and muon parameters were fixed while the $P_f(0)$ parameter was calibrated using the high-Cl CRONUS-Earth samples from Tabernacle Hill and Baboon Lakes.

In the calibration dataset, only whole-rock samples from Tabernacle Hill and hornblende mineral separates from Baboon Lakes had significant production from ³⁵Cl (35-40% and ~75%, respectively). Due to the large difference in the total sample numbers at the two sites (Tabernacle Hill: 24 samples/3 labs and Baboon Lakes: 3 samples/1 lab), the $P_f(0)$ parameter was calibrated at each site individually and then averaged to obtain a representative value. The entire calibration was then performed iteratively to ensure that the spallation, muon, and $P_f(0)$ parameters were all in agreement to threedecimal places. This entire process was repeated for all seven scaling models. Spallation production parameters were influenced only minutely by the changes in muon parameters during calibration iterations, with a similar lack of effect from changing the value of $P_f(0)$.

5. Calibrated Production Rates

5.1. Spallation and low-energy production rates

The final spallation and low-energy production rates from this study are shown in Table 3.

Small differences between the spallation production parameters from Borchers et al. (2015) and the values reported here are because of the inclusion of $P_f(0)$ in the calibration in this study, whereas it was a fixed parameter in Borchers et al. (2015), as well as a small correction to the attenuation lengths used for the samples. The differences between the spallation parameters in the two studies range from 0.2-1.0% across all scaling models. This relative insensitivity is expected owing to the use of low-chlorine surface samples for the spallation calibration, making it relatively insensitive to different values of $P_f(0)$ and attenuation length.

Scaling	Ca at ^{36}Cl (g Ca) $^{-1}$ yr $^{-1}$	K at ^{36}Cl (g K) $^{-1}$ yr $^{-1}$	$P_f(0)$ neutrons (g air) $^{-1}$ yr $^{-1}$
LSDn (Sa)	56.0 $\pm$ 4.1	155 $\pm$ 11	759 $\pm$ 180
LSD (Sf)	56.3 $\pm$ 4.6	153 $\pm$ 12	743 $\pm$ 179
St	52.2 $\pm$ 5.2	150 $\pm$ 15	696 $\pm$ 185
Lm	51.7 $\pm$ 4.9	151 $\pm$ 14	691 $\pm$ 186
Li	60.1 $\pm$ 8.2	142 $\pm$ 19	779 $\pm$ 183
Du	55.0 $\pm$ 12.9	128 $\pm$ 30	720 $\pm$ 182
De	55.6 $\pm$ 13.4	127 $\pm$ 31	722 $\pm$ 181

Table 3- Spallation and $P_f(0)$ production rates with uncertainties. Uncertainties calculated according to the method presented in Section 5.3. See Section 2.1 for definitions of scaling abbreviations.

A statistical test to determine goodness-of-fit was also performed and the resulting p-values are shown in Table 4. Very small values, < 0.05 (Aster et al., 2012), indicate a poor fit to the calibration data. A p-value is reported for each individual step of the calibration. All the calibration steps pass the p-test for all scaling models except for Baboon Lakes, which fails for all models, and potassium spallation, which fails for all models except the LSD and LSDn models.

	LSDn	LSD	St	Lm	Li	Du	De
Spallation Ca	0.829	0.829	0.831	0.832	0.824	0.825	0.824
Spallation K	0.153	0.118	0.017	0.025	0	0	0
Muon Ca	0.767	0.769	0.767	0.785	0.770	0.781	0.773
Muon K	0.714	0.714	0.716	0.721	0.701	0.701	0.698
$P_f(0)$ -TAB	0.205	0.222	0.177	0.184	0.278	0.280	0.281
$P_f(0)$ -BL	0.008	0.008	0.006	0.006	0.012	0.013	0.013

Table 4-Calibration p-values for the various parts of the calibration and different scaling models. Very small values (<0.05) indicate a poor fit to the data. Values listed as zero are smaller than 1×10^{-6} . See Section 2.1 for definitions of scaling abbreviations.

In order to quantify uncertainties directly from the calibration, the statistical methods require that the degree of scatter of the model output be internally consistent with the uncertainties assigned to the input parameters, as assessed by the p-test. Although most of the ^{36}Cl spallation calibrations technically passed the p-test, the small number of sites available for each target element means that the statistical uncertainties produced by the calibration code cannot be assumed to represent a statistically valid estimate of the actual parameter uncertainties and so are not reported here. Furthermore, when calculated for each site individually, the production parameters show some degree of site-to-site variability, which is demonstrated most clearly by the results of the ^{10}Be calibration due to the large number of samples and sites (Borchers et al., 2015; Phillips et al., 2015, this issue-a). In this study, the site-to-site variability is evident primarily in the $P_f(0)$ calibration (Marrero, 2012). The secondary dataset was used to empirically estimate the parameter uncertainties, as described in Section 5.3, and it is these final uncertainties that are presented in Table 3.

5.2. Muon Production Rates

In the Heisinger muon model (Heisinger et al., 2002a; Heisinger et al., 2002b), two nuclide-specific parameters are required to define muon production: σ_0 (fast muon production) and f^* (slow muon absorption). These values have been experimentally determined for some production pathways (e.g. ^{10}Be from SiO_2 , ^{36}Cl from Ca) using irradiation experiments. These accelerator-based experimental values do not always agree with geological data. The Heisinger muon model of ^{10}Be production at depth is higher by a factor of two or more compared to measured values in depth profiles (Braucher et al., 2011; Phillips et al., 2015, this issue-a). To avoid similar problems for muon production of ^{36}Cl , the relevant muon production parameters were reparameterized using two previously analyzed ^{36}Cl profiles, one each for Ca and K. The profiles were previously used to calibrate muon production rates (Stone et al., 1998; Evans, 2001), but did not do so in terms of the

Heisinger muon equations. This reparameterization uses the existing calibration profiles in order to find the parameters necessary to employ the Heisinger model.

To calibrate muon production using geologic samples it is necessary to get below the penetration range for neutron spallation (Stone et al., 1998). In both cases, additional parameters, such as attenuation length and erosion rate, were also fitted to the profile data. The details for each site and the corresponding calibration are discussed below. Table 5 summarizes muon production parameters for all seven scaling models.

5.2.1 Potassium muon reparameterization

The potassium muon depth profile was collected from a granite quarry face in Australia (148° 55'E, 34° 02'S, 540 m). Sixteen samples were collected to a depth of 1600 g cm⁻² (~6 m) below the surface. These were analysed by Evans (2001) and the original study is summarized here. This geomorphically stable area is assumed to be in equilibrium with respect to ³⁶Cl concentrations. The surface above the sample area was uneven; mapping indicated ~15 g cm⁻² missing material. Undisturbed nearby surface samples were collected near the quarry in order to confirm this overburden depth as well as to determine the erosion rate. The ³⁶Cl, ¹⁰Be, and ²⁶Al concentrations in the surface samples confirmed an overburden depth of 15 g cm⁻². Three ³⁶Cl samples indicate a steady-state surface erosion rate of 2.64-7.91 mm kyr⁻¹, depending on the geomorphic location of the sample (e.g. top of slope vs. hillside). Evans (2001) also fitted an erosion rate of 8.5 mm kyr⁻¹ to the depth profile, which is in agreement with the surface sample erosion rate of 7.91 mm kyr⁻¹. The average density of the profile samples from ten measurements was calculated to be 2.643 g cm⁻³. The complete set of original measurements from this core are included as a page in the supplemental spreadsheet.

In this study, the two muon parameters, σ_o and f^* , as well as erosion rate and attenuation length, were simultaneously fitted to the profile data. The calibrated erosion rate and attenuation length were 1.80 ± 0.14 g cm⁻² kyr⁻¹ (6.85 ± 0.53 mm kyr⁻¹) and 165 ± 10 g cm⁻², respectively. The erosion rate agrees well with that of the original study (8.5 mm kyr⁻¹). The values for the muon parameters were $9.4 \pm 12.3 \times 10^{-30}$ cm² for $\sigma_{o,K}$ and $5.8 \pm 1.7 \times 10^{-2}$ for f_K^* . With a p -value of 0.71, the

fit to the data is good, but the uncertainty on $\sigma_{0,K}$ is larger than the calibrated parameter itself indicating that the data does not constrain that parameter well; a deeper profile might allow for better separation of fast and slow muon production. Figure 2 shows the predicted concentration using these calibrated parameters (grey line), and the measured values (points).

{Figure 2: This figure is to be black & white in printed copy and color online. 1.5 or 2-column width. Color and black & white versions of this figure are provided in separate files. Contact author if higher-quality image is required. }

Figure 2-Comparison of the measured sample ^{36}Cl concentrations to those predicted by the calibrated muon model for the potassium calibration profile, shown on a linear plot (a) and a log plot (b). Sample measurement uncertainties are plotted, but are smaller than the points in most cases. The modern production rates for each reaction category are shown on a log plot in (c), as calculated by CRONUScalc using the calibrated model. There was no published fast muon potassium production parameter provided in Heisinger et al. (2002b) so no comparison is presented.

5.2.2 Calcium muon reparameterization

The calcium depth profile, published in Stone et al. (1998) and summarized here, was collected from the wall of an abandoned quarry in the southeastern highlands of Australia (149° 58'E, 34° 19'S, 620 m). The site is geomorphically stable, with a reasonable assumption of steady-state conditions for ^{36}Cl concentrations. The erosion rate could not be determined by independent methods and quarry activity made it difficult to determine if overburden was present. By using nearby surface samples from undisturbed outcrops, the erosion rate was inferred to be 20 mm kyr^{-1} and the overburden depth was calculated to be 174 g cm^{-2} . The density was measured as 2.68 g cm^{-3} and the composition was determined to be almost pure carbonate ($> 99.8 \text{ weight } \%$). The samples were taken from the surface down to a depth of 5400 g cm^{-2} in marble ($\sim 20 \text{ m}$). The shielding was calculated by the original authors using the topography of the area as well as the 20° slope of the hillside. The complete set of original measurements from this core is included as a spreadsheet in the supplemental material.

In this study, the newly calibrated erosion rate of $7.0 \pm 1.0 \text{ g cm}^{-2} \text{ kyr}^{-1}$ ($26 \pm 4 \text{ mm kyr}^{-1}$) and attenuation length of $162 \pm 22 \text{ g cm}^{-2}$ are both very similar to the values from Stone et al. (1998) of 26 mm kyr^{-1} for the fitted erosion rate and the 170 g cm^{-2} assumed for the attenuation length in the original study. Additionally, the additional overburden was calculated to be $140 \pm 23 \text{ g cm}^{-2}$, as compared to the value of 174 g cm^{-2} calculated in the previous study. The values for the fitted muon parameters were $8.3 \pm 1.8 \times 10^{-30} \text{ cm}^2$ for $\sigma_{0,Ca}$ and $1.4 \pm 0.3 \times 10^{-2}$ for f_{Ca}^* . While the fit to the data is good (p -value of 0.77) and the uncertainties are smaller than for the potassium muon parameters, the fitting of another parameter (additional overburden) increases the uncertainty in the calibration. Figure 3 shows the comparison between the measured values in the depth profile and the Heisinger-predicted values using both the original and the newly calibrated values. The newly calibrated parameters improve the match to the measured points. In this profile, the Heisinger parameters lead to an overestimate of production at depth that is more than three times larger than the measured value.

{Figure 3: This figure is to be black & white in printed copy and color online. 1.5 or 2-column width. Color and black & white versions of this figure are provided in separate files. Contact author if higher-quality image is required. }

Figure 3-Comparison of the measured sample ^{36}Cl concentrations to those predicted by the calibrated muon model for the calcium calibration profile, shown on a linear plot (a) and a log plot (b). For comparison, the predicted profile was also calculated using the original Heisinger parameters (dashed lines). Sample measurement uncertainties are plotted, but are smaller than the points in most cases. The modern production rates for each reaction category are shown on a log plot in (c), as calculated by CRONUScalc using the calibrated model. The Heisinger total production (dashed line) reflects the total production rate using the CRONUScalc production model, but using the original Heisinger muon parameters. The profiles are not smooth due to the varying chemical composition of the samples and to a smaller degree the different attenuation lengths of fast and slow muons.

Scaling Model	Ca		K	
	σ_0	f^*	σ_0	f^*

	(10^{-30} cm^2)	(10^{-2})	(10^{-30} cm^2)	(10^{-2})
LSDn (Sa)	8.3 ± 1.8	1.35 ± 0.27	9.4 ± 12.3	5.8 ± 1.7
LSD (Sf)	8.3 ± 1.8	1.36 ± 0.27	9.4 ± 12.3	5.7 ± 1.7
St	8.3 ± 1.8	1.36 ± 0.28	9.3 ± 12.4	5.8 ± 1.7
Lm	8.4 ± 1.8	1.42 ± 0.28	8.9 ± 12.6	6.1 ± 1.7
De	8.3 ± 1.8	1.37 ± 0.28	10.1 ± 11.9	5.2 ± 1.6
Du	8.3 ± 1.8	1.40 ± 0.28	9.9 ± 12.0	5.4 ± 1.6
Li	8.3 ± 1.8	1.40 ± 0.27	10.0 ± 11.9	5.3 ± 1.6

Table 5: Muon production parameters for ^{36}Cl . See Equations 2 and 3 for definitions of parameters.

5.3. Uncertainty on calibrated parameters

Owing to the small number of calibration sites relative to the large number of production parameters, the calibration of the primary dataset did not provide a reliable means for estimating the uncertainty associated with the production rate parameters. The primary dataset provides an unrealistically low estimate because (1) the number of sample sites relative to the number of nuclides calibrated is very low, and (2) the scaling is fitted to those same limited number of sites, so any errors in the scaling method are not accounted for. An alternative method using the distributions of the normalized sample ages for the secondary dataset was used to calculate uncertainties for the calibrated parameters. The secondary dataset, with approximately 12 sample sites, none of which overlap with the primary sites, should more adequately represent the effects of any errors in scaling. However, it provides a conservative estimate of the production rate uncertainty because the data in the secondary dataset are of systematically lower quality than the primary dataset. This method produces an empirical estimate of the production rate uncertainty rather than one statistically derived from the calibration. All uncertainties discussed for the sample parameters and production rates are one-sigma unless otherwise stated.

The observed variability (σ_{obs}) of the sample ages when compared to their independent ages (as quantified by the root-mean-squared error (RMSE) of the calculated ages) was

assumed to be the result of three sources of variability: (1) uncertainty in the assigned independent age (σ_{ia}), which is determined from the bracketing radiocarbon ages and their uncertainties, (2) internal uncertainties (uncertainties on the measured parameters that are propagated in the CRONUScalc program, including uncertainty contributed by the ^{36}Cl analyses, other geochemical analyses, sample location, and surface erosion) and (3) additional ‘unattributed’ sources of uncertainty. This third source includes uncertainties in the production rates as well as other uncertainties that can be difficult to quantify, such as sample thickness on uneven fragments, boulder geometry and neutron loss, exhumation, or forest shielding. The largest contributor to the unattributed uncertainty is probably the inadequacy of the scaling model (including temporal scaling), but it could also include ‘additional’ uncertainties from (1) and (2) above if those uncertainties are underestimated (i.e., if analytical or independent age uncertainties are underestimated, the difference between the estimate and actual value will be in the unattributed category). To ultimately provide ages that reflect a realistic estimation of uncertainty, we assign all of the ‘unattributed’ uncertainty to the production parameters and refer to it as production rate uncertainty (σ_{pr}). For more details, please refer to the discussions in Phillips et al. (2015, this issue-a) and Marrero et al. (2015, this issue).

These three sources of uncertainty are represented in Equation 5, which is based on the form used for standard error propagation (Bevington and Robinson, 1992).

$$(\sigma_{int}^2 + \sigma_{ia}^2 + \sigma_{pr}^2)^{1/2} = \sigma_{obs} \quad (5)$$

The observed variability is different for samples with different dominant production pathways. The samples were sorted into four groups containing approximately equal numbers of samples based on the percent of production resulting from the low-energy production pathway. Spallation results for K and Ca were not examined independently. For each group of samples, we calculated the parameters for observed variability, independent age, and internal uncertainty from the secondary dataset and then we solved Equation 5 for the production rate uncertainty. The calculation methods are described below.

Observed variability was found by calculating ages for each of the samples using the final calibrated production rates. These were converted to normalized ages by dividing the calculated age by the independent age (i.e., completely accurate results would have a normalized age of 1.0). The RMSE was then calculated for each Cl-percent-production group in the secondary dataset (i.e., based on the difference between unity and the actual normalized sample age). The RMSE for each group (Column D, Table 6) was used as the observed variability. The RMSE increases in a fairly linear fashion with increasing percent production from Cl, as seen in Figure 4.

{Figure 4: This figure is to be black & white in all versions. 1-column width. This figure is also provided in a separate file. Contact author if higher-quality image is required. }

Figure 4: Plot illustrating the dependence of the RMSE of the secondary dataset as a function of percentage production from CI, calculated using LSDn scaling. The numbers in parenthesis are the numbers of samples in each production group. The values are given in Table 6.

	A Avg % prod from CI	B Range of % prod from CI	C Number samples	D RMSE norm ages	E σ_{ia} (indpt age uncert)	F RMSE minus σ_{ia}	G σ_{int} (internal uncert)	H σ_{pr} (prod rate uncert)
Secondary	5.1	1-10	16	0.096	0.022	0.093	0.056	0.074
	25.1	10-35	24	0.169	0.025	0.167	0.080	0.147
	49.6	35-70	27	0.171	0.027	0.169	0.082	0.147
	98.9	90-100	12	0.267	0.028	0.266	0.119	0.238
Calib-MS	1.8	0-8	19	0.054	0.015	0.052	0.055	0.000
Calib- $P_f(0)$	41	24-80	17	0.102	0.017	0.101	0.053	0.086

Table6: Data for calculation of sample variation as a function of the percent of the ^{36}Cl production that was from CI, for ages calculated using the LSDn scaling method. See text for detailed description.

The units are normalized age. Results provided for the secondary dataset as well as the calibration dataset, which has been divided into the spallation mineral separates (Calib-MS) and the high-CI

samples used to calculate $P_f(0)$ (Calib- $P_f(0)$). The zero value for σ_{pr} for the mineral separate samples indicates that the analytical uncertainties were likely overestimated. Note: there are no samples in the 70-90% interval for production from CI.

The independent age uncertainty (σ_{ia}) was calculated from the radiocarbon bounds and their uncertainties for samples in the group. These values were relatively small compared to the other uncertainties, ranging from 2.2 to 2.8% (Column E, Table 6). These values are subtracted in quadrature from the observed variability and the results are given in Column F of Table 6.

The internal uncertainty (σ_{int}) was calculated by propagating uncertainties for all input parameters with the production rate uncertainties intentionally set to zero. These were normalized in the same method as that used to normalize the sample ages. The internal uncertainties were averaged for each group and is shown for each category in Column G, Table 6.

The production rate uncertainty (σ_{pr}) was calculated for each group of samples in the secondary dataset, as well as for the spallation and $P_f(0)$ calibration datasets for comparison (Column H, Table 6). The production rate uncertainty associated with the two spallation-production parameters (for K and Ca) were calculated based on the low-Cl sample group (i.e., <10% production from Cl). We do not have a basis to differentiate the fit of the two spallation production rates, thus they are assigned the same uncertainties. For $P_f(0)$, the uncertainty was based on the highest-Cl sample group (85 to 100% production from Cl). To verify these results, the ages for each group were recomputed with the calculated values of the production-rate uncertainty (the value for the production rate itself was left unchanged). The mean of the normalized computed age uncertainties equaled the original observed variability (after the subtraction of σ_{ia}) of the ages in each group of the secondary dataset.

Uncertainties calculated for other scaling methods are presented in Table 3. Although the same uncertainty calculation method was used for all scaling models, not all result in low uncertainty for the spallation parameters. This is due to the poor fit of the scaling model to the calibration dataset in those cases (Borchers et al., 2015).

Given an adequate number of sites and samples in the production groups in the secondary datasets, the use of the production-parameter uncertainty calculated in this way should provide conservative and realistic “external” uncertainties for computed ages. See Phillips et al. (2015, this issue-a) for additional discussion of the uncertainty estimation procedure.

The uncertainties reported here are used in the CRONUScalc program, in both the online surface exposure age calculator as well as the downloadable code (online calculator: <http://web1.ittc.ku.edu:8888/>; downloadable repository: <https://bitbucket.org/cronusearth/cronus-calc>). The calculator returns two types of uncertainties: internal and external. Internal uncertainty includes all the sample measurement uncertainties that are propagated from the sample input provided by the user. The production rate uncertainty discussed here is incorporated only into the external uncertainty calculation where it is added to the internal uncertainty to provide a more realistic, total uncertainty on the sample age. Internal uncertainties are useful for local comparisons using a single nuclide (e.g., retreat rate in a particular valley), but full external uncertainties should be used when comparing exposure ages to results from other nuclides or other chronological techniques (Balco et al., 2008).

This assessment provides the first opportunity to present realistic uncertainties on ^{36}Cl exposure ages, based on the analysis of a large dataset with independent sample ages and covering a broad range of sample compositions. Exposure ages for the primary and secondary datasets were determined with both internal uncertainties and external uncertainties (including production-rate uncertainty). The results show that the increase in uncertainty was between 4-20%. In most samples, this additional uncertainty from the production-rate parameters is some combination of the uncertainty on the spallation and $P_f(0)$ parameters. Although the uncertainty on $P_f(0)$ is large, few samples will see the full magnitude of propagated uncertainty in this range because few samples have >40% production from ^{35}Cl . Most external sample uncertainties remain relatively low (~10%). Although this is larger than the commonly presented internal uncertainties of 3-6% that are largely determined by the analytical AMS uncertainty, the external uncertainties based on evaluation of the secondary dataset represent a more realistic estimate of the accuracy of ^{36}Cl exposure ages.

6. Discussion

6.1. Comparison to previously published production rates

The production rates for this study for all three main pathways are plotted in Figure 5 and compared to those from the previous studies listed in Table 1. Note that the St (and not LSDn)

production rates are plotted so as to be consistent with previous calibrations. The K production rate produced by this study (150 ± 15 at ^{36}Cl (g K) $^{-1}\text{yr}^{-1}$) agrees with the recent production rate study by Schimmelpfennig et al. (2014) of 148 ± 8 at ^{36}Cl (g K) $^{-1}\text{yr}^{-1}$ as well as those of Phillips et al. (2001) and Evans et al. (1997) calibrations. The two studies that are not consistent are the Schimmelpfennig et al. (2011) study and the Swanson and Caffee (2001) study. A possible explanation of the former is a problem with the K-Ar independent age of the flow or erosion on the samples (Schimmelpfennig et al., 2014). The Swanson and Caffee (2001) study has been the subject of much debate.

{Figure 5: This figure is to be black & white in printed copy and color online. 2-column width. Color and black & white versions of this figure are provided in separate files. Contact author if higher-quality image is required. }

Figure 5- Comparison of production rates produced in this study to those found in the previous studies shown in Table 1. All studies are compared using St scaling, including this study. The shaded regions show the range of the production rate plus uncertainty from this study. *Swanson and Caffee (2001) production rates include production from muons, where relevant.

There is significant scatter in the calcium production rates, with this study only overlapping with two previous studies. The use of the same site in both this study and Stone et al.,(1996) likely

contributes to the similar results. We believe that the CRONUS-based result is robust but we cannot explain the discordancy with the entire literature dataset. Although the Swanson and Caffee (2001) study is well-known to have discrepant results, the remaining discrepancy of 20-27% with the non-overlapping studies indicates that additional calcium calibration sites are needed to investigate the discrepancy and better constrain the production rate.

The uncertainty for $P_f(0)$ in this study is larger than for the other production reactions due to the wide scatter in the ages from the high-Cl dataset, which was composed of leachates from high-Cl feldspars by Evans et al. (1997) and quartz samples from Scotland by Evans (2001). This causes the value from this study to be within uncertainty of all previous values determined for $P_f(0)$. However, even if the uncertainty was half of that calculated, it would still overlap within uncertainty of the other calibrated values. The nominal value ($696 \text{ n (g air)}^{-1} \text{ yr}^{-1}$) agrees best with the value produced by Evans(2001) ($740 \text{ n (g air)}^{-1} \text{ yr}^{-1}$), although it is in agreement with the slightly lower value ($626 \text{ n (g air)}^{-1} \text{ yr}^{-1}$) of Phillips et al. (2001) as well as the two other studies that produced higher, but relatively similar, values of $P_f(0)$ of $760 \text{ n (g air)}^{-1} \text{ yr}^{-1}$ (Alfimov and Ivy-Ochs, 2009) and $762 \text{ n (g air)}^{-1} \text{ yr}^{-1}$ (Swanson and Caffee, 2001). There are a considerable number of obstacles to determining $P_f(0)$ in a geological calibration, primarily its dependence on whole-rock composition and sensitivity to hydrogen. Current production models may not adequately represent all of the processes potentially acting on production by low-energy neutron absorption. Although the uncertainty on $P_f(0)$ is larger than the other pathways, the calibrated value from this study represents the best value possible from the CRONUS-Earth calibration samples; in the following section we evaluate the performance of $P_f(0)$ and the other calibrated rates for a variety of samples. If low uncertainties are required, low-Cl samples (such as mineral separates) are still recommended until the reasons for the variability in $P_f(0)$ are resolved.

6.2. Assessing the calibrated production rates

Figure 6 shows the scatter of the normalized ages of the primary calibration dataset as a function of latitude, elevation, and age. There do not appear to be any clear trends, although both the Baboon Lakes and Scotland sites plot to one side of the line; this could just simply be under-

sampling. Unfortunately, the primary calibration dataset is too small to be used to assess possible bias in the production rates as a function of target-element composition or other factors. Instead, we can use the secondary dataset for this purpose.

{Figure 6: This figure is to be black & white in printed copy and color online. 2-column width. Color and black & white versions of this figure are provided in separate files. Contact author if higher-quality image is required. }

Figure 6-The normalized ages (calculated age divided by independent age) for the primary calibration dataset (including the samples used to calibrate $P_f(0)$) shown plotted by (a) site (from left to right: Tabernacle Hill whole-rock, Baboon Lakes hornblende, Tabernacle Hill Ca-rich plagioclase mineral separates, Huancané K-feldspar mineral separates, and Scotland K-feldspar mineral separates), (b) latitude, (c) elevation, and (d) independent age. Uncertainties are only shown in panel (a) for simplicity, but are the same for all other plots in this figure.

A similar comparison of latitude, elevation, and independent age was performed using the secondary dataset, a completely independent dataset that was not used for calibration. The results are shown in Figure 7. Most sites plot evenly around one and are internally consistent. Site-to-site variability larger than that expected based on inter-lab variability is evident, especially for the New England and Baboon Lakes sites (Phillips et al., 2015, this issue-a). In many cases, the spread within individual sites exceeds the level expected based on analytical uncertainties alone. The latitude and independent age plots show no trend. The elevation plot appears to show decreasing normalized age with increasing elevation. This could be an indication that the problem with the variability is due to problems with scaling. This plot may also be skewed by the geographical proximity of the high elevation sites because they are all from the Sierra Nevada region. The root mean squared error for the ages from entire secondary dataset was 18.4%.

{Figure 7: This figure is to be black & white in printed copy and color online. 2-column width. Color and black & white versions of this figure are provided in separate files. Contact author if higher-quality image is required. }

Figure 7-Secondary dataset plotted by (a) site, (b) latitude, (c) elevation, and (d) independent age.

Note that the data from Evans et al. (1997) is broken into potassium feldspar samples (Evans, 1997-K) and leachate samples (Evans, 1997-Cl). The samples plotted here include Baboon Lakes samples, but only those not used in the calibration. Baboon Lakes samples were not used in the uncertainty analysis. Phillips et al. (2001) and Phillips et al. (2009) are a subset of samples published in the paper. See Appendix 2 for a description of the included samples.

This same secondary dataset was plotted against percent production from ^{35}Cl (Figure 8) to examine the accuracy of the $P_f(0)$ parameter. An ill-fitting value for $P_f(0)$ will manifest itself as a systematic divergence from the line for normalized age equals 1. There is no apparent systematic trend with increasing production from ^{35}Cl , indicating that the $P_f(0)$ value performs reasonably well over the range of the entire secondary dataset, including high-chlorine samples. An alternative calibration of $P_f(0)$ employing chi-squared minimization on the secondary dataset, yielded a minimum

chi-squared between 700 and 750 neutrons (g air)⁻¹ yr⁻¹, which is in agreement with our calibrated value (Marrero, 2012). For comparison, the secondary dataset is also plotted in the same figure with the $P_f(0)$ values derived from the Tabernacle Hill samples (916 neutrons (g air)⁻¹ yr⁻¹) and the Baboon Lakes samples (601 neutrons (g air)⁻¹ yr⁻¹). Both of these produce age distributions that are correlated with increasing Cl content (positively and negatively, respectively), indicating that the value from the primary dataset calibration is closer to the actual $P_f(0)$ than alternative calibrations based on either of these sites alone.

{Figure 8: This figure is to be black & white in printed copy and color online. 1-column width. Color and black & white versions of this figure are provided in separate files. Contact author if higher-quality image is required. }

Figure 8- The normalized (calculated/independent) ages of the samples in the secondary dataset. The dataset is plotted using three different values of $P_f(0)$ to demonstrate the potential changes in sample ages. Linear trendlines are shown for each dataset.

6.3. Reasons for variability in $P_f(0)$

The $P_f(0)$ values calibrated from the two higher-Cl sites (BL and TAB) by themselves produced biased age distributions with increasing Cl content. Employing the LSDn scaling method, the Baboon Lakes high-Cl mineral separates (three samples) produces a $P_f(0)$ of $601 \text{ n (g air)}^{-1} \text{ yr}^{-1}$. Tabernacle Hill whole-rock samples from three labs (24 samples total) produces a $P_f(0)$ value of $916 \text{ n (g air)}^{-1} \text{ yr}^{-1}$.¹ There is no a-priori reason to prefer one of these sites over the other. The Baboon Lakes site has the advantage that the preponderance of the production is from Cl ($\sim 75\%$), but there are limited analyses. The Tabernacle Hill whole-rock samples are numerous, but only $\sim 40\%$ of the ^{36}Cl production is from Cl. These two values were averaged to obtain a $P_f(0)$ value of $759 \text{ n (g air)}^{-1} \text{ yr}^{-1}$.

There are two possible explanations for the variability in $P_f(0)$: 1) The low-energy neutron flux is sensitive to the water content of the environment, especially snow overlying the rock surface (Dunai et al., 2014; Zweck et al., 2013). Water content differs between the two sites, with Baboon Lakes being at high altitude with the potential for snow cover for more than six months of the year and Tabernacle Hill being located in a semiarid setting with probably infrequent snow cover. Inability to completely model the effects of differing water content (especially snow) may introduce a bias. Alternatively, 2) the variability is caused by the same processes that Borchers et al. (2015) hypothesized (scaling framework or site characterization) caused excessive site-to-site variability of the production rates calculated for other nuclides in the CRONUS calibration. Similar, although generally smaller, site-to-site variability was observed by Borchers et al. (2015) in other nuclides (^{10}Be , ^3He) during the production rate calibration, indicating the presence of a process that is either not included in the model or is being modelled incorrectly. If the processes causing spallation are improperly modelled, the model then compensates by requiring a different value for $P_f(0)$.

The second hypothesis can be assessed by looking at the divergence of the normalized ages determined by other nuclides (Table 7). If the issue is scaling or site characterization then we would expect all nuclides to be likewise affected. The site-to-site variability in the overall calibration is not limited to sites with high-Cl samples, and while it is difficult to rigorously test these two hypotheses, it seems that the ^{36}Cl does follow the same trend as the other nuclides. Incorrect independent ages would also be expected to affect all nuclides at a particular site, although it is more likely that there is a scaling problem than significant independent age problems with multiple carefully vetted sites. The observed site-to-site variability trends do lend credence to the hypothesis that our scaling models do not capture all the physical processes affecting cosmogenic nuclide production.

Site	^{36}Cl	Other Nuclide
TAB	o(+)	o(+) ^3He
SCOT	++	+ ^{10}Be
BL	–	o(-) ^{10}Be
NE	+	+ ^{10}Be
PERU	o	o(-) ^{10}Be

Table 7-Comparison of site-to-site variability across nuclides. Subjective degree of deviation from the 1:1 ratio of measured concentration/predicted concentration, as calculated in Borchers et al., (2014). Rankings from most positive deviation to most negative: ++, +, o(+), o, o(-), -, -. The 'o' indicates that a number of samples from that site overlap the 1.0 normalized age line and indicators after that indicate if there was a bias in either direction.

For any sample where ^{36}Cl production from ^{35}Cl is significant, the composition parameters that determine the incoming low-energy neutron flux can affect the final result. The major and minor element composition of samples are typically well-determined via XRF, ICP, or NAA. However, water content at the time of sampling is almost never measured and this parameter can vary through time. Depending on composition, water content can be critical to the correct calculation of the incoming neutron flux. Neutrons are strongly moderated by water (Zreda et al., 2008), which changes not only the intensity of the flux but the incoming energy range as well. Dunai et al. (2014) described an increase in the thermal neutron flux by a factor of up to 2.5 from the presence of very small amounts of water (2-3 cm of water equivalent) as a layer over the rock surface. A similar effect is

seen in the snow shielding simulations performed by Zweck et al. (2013) where they show modelling results that environmental water (e.g., lake, vegetation), even at some distance from the sample, may be able to affect the low-energy neutron flux and consequently the production rate. The use of low-energy neutron counters to determine soil-water content over a footprint of 600 m also lends credence to this idea (Desilets and Zreda, 2013; Zreda et al., 2008).

In the calibration, the Baboon Lakes high-Cl mineral separate samples would be most affected by the effects of hydrogen. When we tested the sensitivity of the calibrated value for $P_f(0)$ by incorporating the snow-shielding factors of Zweck et al. (2013), the site-specific value of $P_f(0)$ increased by only 1.1%. Located in the desert, the samples from Tabernacle Hill are unaffected and the site-specific value of $P_f(0)$ does not change, so the overall value of $P_f(0)$ is impacted by only half this amount when the final value is calculated. We do not consider snow shielding at Baboon Lakes to be an issue.

6.4. Sensitivity Analysis

A sensitivity analysis was performed to determine the influence of input parameters on final results. This was accomplished by calculating partial derivatives for each parameter with respect to age, while holding all other parameters constant. A wide assortment of the calibration samples was used in an attempt to include differing compositions and locations. The derivatives were calculated for each parameter in terms of kyr age change per one unit change in the parameter. For each sample, the derivative was then multiplied by a reasonable uncertainty for that particular parameter. The parameters and their assigned uncertainties are shown in Figure 9. These are conservative uncertainties designed to test the broadest possible impact on any given sample.

The samples were divided into four categories and the major, minor, and non-contributing factors were determined for each group. The results are summarized in Figure 9. For the surface samples, the major factors contributing to the uncertainty on the age, arbitrarily defined as parameters that have more than a 2.5% effect on sample age, are the nuclide concentration and average atmospheric pressure. Uncertainties on nuclide concentrations can be reliably estimated, whereas the pressure is an

unexpected major contributing factor. Each of these parameters can contribute up to 8% uncertainty to the age. Minor contributing factors, defined as parameters that affect most of the samples in the group by 1.0 to 2.5%, include erosion rate, shielding, attenuation length, thickness, and density. For high-Cl samples, such as whole-rock basalts, the composition parameters are also minor contributing factors, although the degree of sensitivity varies significantly depending on composition and environmental conditions. The lack of sensitivity to rock water content should not be assumed to apply equally to all rock types. This sensitivity analysis does not include the effects of environmental water (e.g. water in the soil surrounding the sample) that may affect production rates.

This sensitivity analysis assumed that the uncertainties reported by the analysing labs (e.g. XRF, ICP) was correct. However, this does not account for differences originating from non-quantitative sample splitting or differences between XRF labs. For example, the TAB samples were split and sent for Cr analysis by two different labs (DAL and UW). The results produced by the two XRF labs were different, with the lab used by DAL producing values that were sometimes twice those reported by the lab used by UW. Although Cr content is only a minor contributing factor in the sensitivity study, the relatively high Cr contents of the sample (200-400ppm) and the large discrepancies between the labs turns this into a potentially significant contributing factor. The $P_f(0)$ calibration samples used the average of the two concentrations for all the whole-rock samples, regardless of lab. In order to test real-world sensitivity to Cr concentrations, the TAB $P_f(0)$ value, was calculated using the UW (low) and DAL (high) Cr values and compared to the average value actually used in the calibration. The high-Cr, site-specific TAB $P_f(0)$ value is 4% higher than the average value, leading to a 2% higher total value of $P_f(0)$. This is well within the specified uncertainty of the parameter, but it demonstrates the importance of quantitatively splitting homogenous material, the necessity of measuring trace elements for high-Cl samples, and the need for a ^{36}Cl intercomparison that includes comparison of measured sample compositions.

{Figure 9: This figure is to be black & white in printed copy and color online. 1.5 or 2-column width. Color and black & white versions of this figure are provided in separate files. Contact author if higher-quality image is required. }

Input	Sample Range	uncert	units	Whole Rock	Min Sep	High Cl - depth	Low-Cl - depth
Conc	Actual	Actual	at/g				
erate	0.05-1.9	0.5*actual	g/cm ²				
water cont	0.5	1	vol %				
density	2.3-2.8	0.3	g/cm ³				
thick	3-8	1	cm				
press	565-974	5	hPa				
shielding	0.98-1.00	0.005	--				
Λ_{eff}	140-170	10	g/cm ²				
Bulk Na ₂ O	3-4	0.4	wt %				
Bulk Al ₂ O ₃	14-19	0.4	wt %				
Bulk K ₂ O	0.5-5	0.4	wt %				
Bulk CaO	0.2-10	0.4	wt %				
Bulk TiO ₂	0.2-2	0.4	wt %				
Bulk MnO ₂	0.01-.15	0.4	wt %				
Bulk Fe ₂ O ₃	1-11	0.4	wt %				
B	0-100	10	ppm				
Sm	2-13	2	ppm				
Gd	2-13	2	ppm				
U	0.3-10	2	ppm				
Th	1-12	2	ppm				
Cr	0-400	20	ppm				
Targ K	0.2-13	0.1	wt %				
Targ Ca	0.2-10	0.1	wt %				
Targ Cl	1-700	0.5	ppm				
Depth	0-400	5 cm equiv.	g/cm ²				

Figure 9-Relative influence of realistic uncertainties on the ages of four types of samples: whole-rock (high-Cl; > 85 ppm) surface samples, mineral separate (low-Cl) surface samples, high-Cl depth samples, and low-Cl depth samples. The dark red boxes indicate major influence (>2.5%), light blue indicates minor influence (1-2.5%), white indicates non-contributing parameters, and hashed pattern indicates that the parameter changes in influence with depth and is a minor contributor. Only parameters that contribute to the uncertainty in some way are shown. All other input parameters are considered “non-contributing” to the uncertainties on samples. The ‘uncert’ represents the actual level of the uncertainty applied in the analysis, with units also shown. ‘Targ’ indicates the target composition. Depth uncertainties of the equivalent of 5 cm were used only for depth-profile samples. ‘Erate’ indicates the erosion rate.

Hypothetical depth-profile samples were created by analysing two surface samples as if they were at depths of 25, 50, 75, 100, 250, 300, and 400 g cm⁻². The results are similar in many ways to the surface sample sensitivity analysis discussed above. Many of the major contributing factors, such as uncertainty in concentration and pressure, still contribute significantly to the uncertainty of the sample. Additional factors also contribute to uncertainty but were not significant in the previous analysis, such as attenuation length, which is more important in depth profiles than surface samples, and depth to the top of the sample. Once again, the difference between high-Cl and low-Cl samples is clear. The high-Cl profile uncertainties depend significantly on the composition parameters and water content, while the spallation-dominated profile uncertainties depend more on the target composition.

The implications of the sensitivity analysis are twofold. First, high-Cl sample results depend significantly on the compositional parameters, especially trace elements and water content. This is an expected result, but it does highlight the importance of measuring these parameters accurately if samples are chlorine-rich. If the samples are known to be low in chlorine, these parameters might be justifiably ignored. Second, mean atmospheric pressure is the single biggest contributing factor other than the measured concentration of the nuclide. As sample pressure is typically derived from elevation measurements, this means that accurate sample elevation is important and some sample exposure ages may be particularly sensitive to the elevation scaling that is used in scaling models. Pressure variability through time is difficult to tightly constrain and the pressure history likely varies from site to site (Staiger et al., 2007). This offers a possible explanation for previously unexplained site-to-site variability. If future work could lead to the reduction of pressure uncertainty, the uncertainties on the ages of the samples could also be reduced. Alternatively, we cannot preclude that other variables may co-vary with elevation (e.g., snow cover, ash cover, vegetation cover) and may have been improperly evaluated as their prehistoric magnitudes are difficult to evaluate accurately.

Not included in the sensitivity analysis is the difference between various calculators, more specifically, the assumptions implicit in available calculators. For low-Cl samples, most sample ages will be very similar regardless of the calculator used. High-Cl samples, such as those presented in Kelly et al. (2008), can vary significantly for reasons such as B concentration or the assumption of

radiogenic equilibrium. For example, the value of the dilute resonance integral for B is listed as 1722 barns (Mughabghab, 2006), however, when the value is corrected for the natural abundance, the value reduces to 343 barns (the default used in CRONUScalc). Both values have been used in various ^{36}Cl calculators and the subtle differences are not always apparent from the calculator interface. Using the Kelly et al. (2008) samples as examples, identical samples run through CRONUScalc using the two different values for the dilute resonance integral for boron differ by 3.3% in age.

Another example of implicit differences in calculators is the assumption of radiogenic equilibrium. CRONUScalc currently assumes that samples are in radiogenic equilibrium with the measured concentrations of U and Th. This is generally true for old rocks, but may or may not be true for younger rocks, such as newly erupted basalts. Alternatively, the Schimmelpfennig et al. (2009) spreadsheet allows for an age of formation to be entered for the rock and radiogenic production is calculated from that point onwards. This only applies to samples with significant production from the low-energy pathway and for most samples the difference between these two assumptions is generally negligible, but in the case of very high-Cl samples, such as the Kelly et al. (2008) samples, with relatively high U (average 7.6 ppm) and Th (average 26.1 ppm), the change in age is 15-29%. Further research to differentiate between these two assumptions is necessary to reduce uncertainties for high-Cl samples. While these samples represent a rare combination of conditions, it is worth noting that there may be other samples that fall into these (or other unidentified) categories that result in ages that are significantly different, despite relatively similar production mechanisms elsewhere.

7. Conclusions

This study completes the calibration presented in Borchers et al. (2015) by including the muon reparameterization, calibration of $P_f(0)$, and determination of the uncertainties associated with the production-rate parameters, thus providing a single, coherent set of production-rate parameters for ^{36}Cl from the CRONUS-Earth dataset. Based on the increasing body of evidence that variations in water content can cause significant variations in the production rates of ^{36}Cl from low-energy reaction neutrons (Zweck et al., 2013; Dunai et al., 2013) that are not accounted for by the classical neutron-diffusion equations (Phillips et al., 2001), ^{36}Cl users should be cautious regarding the interpretation of

high-chlorine content (>80 ppm Cl) samples. The observation of significant site-to-site variability in the calibrated $P_f(0)$ parameter does indicate that there is variability that is not currently explained by the CRONUScalc model. It may be significant that when the classical equations and production rates from this study were tested against the secondary dataset, they performed in a consistent fashion even for samples with a significant production from Cl. Although the observed variability of the normalized ages increased significantly, the high chlorine ages still exhibited a mode near unity and a distribution close to normal.

This study has also identified several steps that would be useful for future improvement of cosmogenic ^{36}Cl techniques. A factor that we have not included in our deliberations are inter- and intra-lab variability. These latter issues are complex and encompass issues ranging from Cl extraction techniques to analytical determinations of target chemistry. Future studies should incorporate robust tests for determining the impact of laboratory issues on ^{36}Cl production rates. Additional sites of varying ages and geographic locations would be immensely useful. The continuing difficulty in determining $P_f(0)$ needs to be systematically investigated because the use of samples containing significant Cl cannot be eliminated while maintaining the practicality and versatility of ^{36}Cl . Although $P_f(0)$ would also benefit from additional calibration sites, studies to verify assumptions (such as radiogenic equilibrium) or investigating the effect of environmental water would improve the fundamental theory and reduce the uncertainty on that parameter. Data arising from the increasing use of low-energy cosmic-ray neutrons for monitoring environmental water content (Zreda et al., 2012) may enable improvement of the theory for modelling the production of ^{36}Cl by low-energy neutrons.

The production rates introduced here have been incorporated into the CRONUScalc program, which is available as an online surface-exposure age calculator (<http://web1.ittc.ku.edu:8888>) or available to download from the open-source repository (<https://bitbucket.org/cronusearth/cronus-calc>) in order to run calibrations or perform other tasks. As additional calibration sites are found to constrain the production rates, the calibration will need to be revisited in order to improve the production rates.

Statement of Contributions

Shasta Marrero participated in field sampling and sample preparation and analysis of samples from Tabernacle Hill, Scotland, and Copper Canyon, sample preparation of the Quelccaya samples, co-wrote the data-reduction software, and performed the actual calibration. Fred Phillips oversaw the project, participated in sampling at Tabernacle Hill, Scotland, and Copper Canyon, and led the sampling effort at Quelccaya. Marc Caffee participated in sampling at Tabernacle Hill and Scotland and prepared and analyzed samples from those sites, as well as overseeing all ^{36}Cl analyses performed at PRIME Lab. John Gosse prepared samples from Tabernacle Hill. All authors participated in writing and revising the manuscript.

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Appendix 1: Additional information about calibration samples

Table A1- Location information (latitude, longitude, elevation, pressure), shielding, erosion rates, and the ^{36}Cl concentrations for the samples in the calibration dataset that are not already published elsewhere. The pressure was calculated using the ERA-40 reanalysis as programmed in CRONUScalc (Marrero et al., 2015, this issue). The depth to top of sample is 0 for all samples. Uncertainties were estimated as follows: elevation ± 10 m; thickness ± 0.5 cm; and shielding ± 0.005 . Some parameters and uncertainties varied by site. For Peru, bulk density was measured as $2.29 \pm 0.14 \text{ g cm}^{-3}$; samples were collected in 2008; and water content is assumed to be 2% (entered as 0.02 in CRONUScalc). For Scotland, bulk density was measured as $2.55 \pm 0.20 \text{ g cm}^{-3}$; samples were collected in 2006; and water content is assumed to be 0%. For Baboon Lakes, bulk density was measured as $2.75 \pm 0.10 \text{ g cm}^{-3}$; samples were collected in 2007; and water content is assumed to be 0.5% (entered as 0.005 in CRONUScalc). λ_{eff} is the effective attenuation length. Effective attenuation length was calculated using the table in Marrero et al. (2015, this issue), which has been updated and may provide

different values than those previously published for these samples. The full dataset can be found in the supplemental spreadsheet.

Name	Latitude	Longitude	Elev	Press	Thick	Shielding	E-rate	λ_{eff}	^{36}Cl conc.
-	dd	dd	m	hPa	cm	--	mm/kyr	g/cm^2	10^4 atoms/g
HU08-01-SAN	-13.9449	289.1046	4854	566	7	0.997	8.1±3.2	182	164.5±7.7
HU08-02-SAN	-13.9464	289.1076	4862	566	5	0.999	4.8±1.6	186	166.2±7.9
HU08-03-SAN	-13.9461	289.1073	4859	566	6	0.995	3.2±1.6	180	171.7±8.1
HU08-04-SAN	-13.9455	289.1072	4848	567	5	0.991	8.1±3.2	177	146.3±7.0
HU08-06-SAN	-13.9445	289.1052	4843	567	7	0.999	4.8±1.6	188	157.2±7.6
HU08-10-SAN	-13.947	289.1131	4860	566	3.2	0.997	4.8±1.6	184	165.4±8.0
HU08-11-SAN	-13.9472	289.1132	4860	566	6	0.999	6.5±3.2	188	164.0±7.8
HU08-14-SAN	-13.9489	289.1144	4871	565	5	0.996	3.2±1.6	189	159.2±7.7
HU08-15-SAN	-13.9484	289.1146	4869	565	6	0.995	6.5±3.2	189	146.0±7.1
HU08-16-SAN	-13.9468	289.1138	4867	565	6	0.997	3.2±1.6	183	152.3±7.5
06-SKY-01-KFSP-NXHN	57.24081	354.0273	442	958	10	0.924	0±0	140	17.64±0.82
06-SKY-06-KFSP-NXHN	57.24249	354.0341	310	974	8	0.99	0.2±0	153	14.12±0.79
06-SKY-07-KFSP-NXHN	57.24269	354.0352	300	975	4	0.992	0.2±0	147	14.71±0.73
06-SKY-08-KFSP-NXHN	57.2425	354.0347	304	974	4	0.988	0.2±0	141	17.6±0.85
BBL07-02	37.1682	241.3796	3387	676	5.5	0.983	1±0.5	161	318±20
BBL07-03	37.168	241.3799	3387	676	4.5	0.989	1±0.5	161	225±12
BBL07-01	37.1683	241.3799	3380	676	4	0.985	1±0.5	161	326±15
Peru									
Scotland									
BL									

Table A2 – Bulk rock composition XRF results for the calibration dataset. Uncertainties on all elements are 0.01 weight percent. All results are directly from XRF, other than analytical water (An. Water) and CO₂, which are estimated by the investigator from the LOI reported from XRF. Tabernacle Hill (WR)-PRIME analyses are averages from several quantitative splits sent for XRF. The values presented in this table are corrected and supersede the values presented in Lifton et al. (2015). The full dataset can be found in the supplemental spreadsheet.

Name	SiO ₂	TiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	An. Water	CO ₂
-	bulk rock weight %											
HU08-01-SAN	74.20	0.21	14.00	1.40	0.01	0.22	0.70	3.20	4.90	0.21	0.90	0.00
HU08-02-SAN	74.18	0.20	13.93	1.50	0.01	0.22	0.72	3.19	4.82	0.20	0.89	0.00
HU08-03-SAN	74.21	0.21	14.35	1.30	0.01	0.23	0.76	3.23	4.85	0.22	0.51	0.00
HU08-04-SAN	74.03	0.20	14.04	1.37	0.01	0.22	0.70	3.11	4.79	0.20	1.22	0.00
HU08-06-SAN	73.27	0.21	14.28	1.40	0.01	0.23	0.75	3.29	4.89	0.21	1.33	0.00
HU08-10-SAN	72.98	0.20	13.87	1.50	0.01	0.22	0.69	3.06	4.93	0.19	2.22	0.00
HU08-11-SAN	73.69	0.20	14.02	1.52	0.02	0.25	0.73	3.12	4.88	0.19	1.25	0.00
HU08-14-SAN	73.38	0.22	14.38	1.45	0.01	0.24	0.72	3.15	4.99	0.18	1.15	0.00
HU08-15-SAN	73.02	0.21	14.20	1.73	0.01	0.25	0.71	3.11	4.95	0.20	1.49	0.00
HU08-16-SAN	68.83	0.21	13.62	1.40	0.02	0.24	0.70	3.10	4.83	0.18	6.75	0.00
Peru												
06-SKY-01-KFSP-NXHN	74.10	0.27	12.70	3.41	0.05	0.08	0.36	4.11	4.99	0.02	0.65	0.03
06-SKY-06-KFSP-NXHN	73.10	0.24	12.40	3.13	0.05	0.04	0.23	4.12	4.95	0.02	0.01	0.01
06-SKY-07-KFSP-NXHN	73.80	0.26	12.60	3.10	0.05	0.06	0.36	3.93	5.07	0.02	0.85	0.03
06-SKY-08-KFSP-NXHN	74.50	0.25	12.60	3.05	0.04	0.04	0.20	3.84	5.19	0.02	0.95	0.05
Scotland												
BBL07-02	59.16	0.82	17.23	7.06	0.11	3.1	5.5	3.87	2.16	0.05	0.85	0
BBL07-03	68.005	0.37	15.12	3.29	0.05	1.06	3.22	3.48	3.48	0.02	1.785	0
BBL07-01	68.4	0.335	16.02	3.32	0.04	1.45	3.86	3.495	3.22	0.05	0.65	0
BL												
05TAB01	49.55	1.50	17.30	10.61	0.16	6.60	9.49	2.98	0.78	0.21	0.65	0
05TAB03	49.13	1.51	17.02	10.90	0.16	6.85	9.49	3.12	0.83	0.26	0.55	0
05TAB04	48.96	1.55	16.64	11.82	0.18	7.67	9.18	3.06	0.69	0.24	0.34	0
05TAB05	49.13	1.61	16.78	11.32	0.17	7.22	9.33	3.06	0.71	0.24	0.29	0
05TAB06	49.09	1.62	16.67	11.56	0.17	7.19	9.35	2.89	0.74	0.28	0.33	0
05TAB07	49.33	1.62	16.60	11.68	0.17	7.18	9.39	2.84	0.87	0.31	0.05	0
TAB (WR)-PRIME												

Table A3 – Trace element composition of the bulk rock for the calibration dataset. All units are parts per million in the bulk rock. The full dataset can be found in the supplemental spreadsheet.

Li(ppm)
430±20
430±40
440±40
440±40
470±40
440±40
450±40
420±40
450±40
430±40
10±3
20±5
20±2
10±2
20±10
20±10
20±10

Name	Cl (ppm)	B (ppm)	Sm (ppm)	Gd (ppm)	U (ppm)	Th (ppm)	Cr (ppm)
HU08-01-SAN	10±10	60±6	3.2±1	2.34±1.0	9.62±0.1	12.40±0.5	100±50
HU08-02-SAN	80±1	60±6	3.2±0.3	2.34±0.2	9.62±0.9	12.40±1.0	140±40
HU08-03-SAN	83±1	160±10	3.2±0.3	2.47±0.2	9.02±0.9	12.50±1.0	50±50
HU08-04-SAN	75±1	70±7	3.1±0.3	2.22±0.2	9.26±0.9	11.40±1.0	50±50
HU08-06-SAN	97±2	80±8	3.4±0.3	2.23±0.2	9.30±0.9	12.00±1.0	60±50
HU08-10-SAN	91±2	100±10	3.1±0.3	2.19±0.2	9.46±0.9	10.60±1.0	100±50
HU08-11-SAN	70±1	120±10	3.1±0.3	2.03±0.2	8.45±0.8	11.00±1.0	110±50
HU08-14-SAN	112±2	100±10	2.9±0.3	1.96±0.2	8.54±0.8	10.70±1.0	50±50
HU08-15-SAN	106±2	90±9	2.9±0.3	2.31±0.2	10.90±1.0	11.20±1.0	130±50
HU08-16-SAN	110±3	120±10	3.1±0.3	1.95±0.2	8.75±0.9	10.70±1.0	50±50
Peru							
06-SKY-01-KFSP-NXHN	39±2	5±5	14.6±1	15.6±1.0	1.85±0.1	10.10±0.5	5±5
06-SKY-06-KFSP-NXHN	7±3	5±5	13.0±1	13.4±1.0	1.88±0.1	10.70±0.5	50±50
06-SKY-07-KFSP-NXHN	11±2	5±5	11.1±1	10.6±1.0	1.98±0.1	10.30±0.5	5±5
06-SKY-08-KFSP-NXHN	23±	5±5	10.1±1	9.41±1.0	2.02±0.1	10.30±0.5	5±5
Scotland							
BBL07-02	296±40	10±10	1.9±0.1	2.09±0.05	0.55±0.05	1.7±0.1	20±10
BBL07-03	85±21	10±10	1.9±0.1	2.09±0.05	0.55±0.05	1.7±0.1	20±10
BBL07-01	102±22	10±10	1.9±0.1	2.09±0.05	0.55±0.05	1.7±0.1	20±10
BL							

Table A4: Cr concentrations for the Tabernacle Hill whole-rock samples. The values measured by the reporting lab (UW) were used for the mineral separate samples (values presented in Lifton et al., 2015) and the average of DAL and UW measurements was used for all the whole-rock samples (DAL, NMT, and PRIME). Li concentrations were changed to 1 ppm for all whole-rock samples. These values supersede the whole-rock values presented in Lifton et al. (2015). The full dataset can be found in the supplemental spreadsheet.

Cr (ppm)	05TAB01	05TAB02	05TAB03	05TAB04	05TAB05	05TAB06	05TAB07
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DAL	340	409	408	405	408	342	407
Average	255.0	304.5	284	302.5	304	271	303.5

Appendix 2: Secondary Dataset Site Descriptions

Site	Use	Notes about outliers and modifications of datasets
TAB-MS	Ca spallation	Samples processed by John Stone in 1996 were added to this dataset (identical location, identical lab, well documented).
TAB-WR	Pf(0)	3 labs contributed data to this site. Several samples were removed as outliers. All removed samples were >2 standard deviations from the mean for that lab.
SCOT	K spallation	One additional site from SCOT for Ca spallation was removed due to the likely presence of inheritance (see Marrero, 2012).
PERU-MS	K spallation	Two sets of mineral separates were produced for this site. The samples from the NMT lab were very inconsistent with the samples from Stone at University of Washington. No definitive cause for the discrepancy was found, but the NMT samples were deemed the likely source of the problem and the UW samples were used (Marrero, 2012).
PERU-WR	Removed	These samples likely suffered from the same bias that caused problems in the PERU-MS mineral separates from NMT as they resulted in physically impossible results for Pf(0) (Marrero, 2012).
Baboon Lakes (BL-B)	Pf(0)	Hornblende mineral separates prepared by M. Hinz at NMT.
Baboon Lakes (BL)	Second	Feldspar mineral separates and whole-rock samples prepared by M. Hinz at NMT. One sample was removed completely: F2BL07-03. The concentration is almost an order of magnitude lower than any of the other samples. The independent age for this site is 13,400 and the age this sample produces is only 700 yrs.
New England	Second	These samples were moved into the secondary dataset due to concerns about the erosion rate and possibility of cover.
Evans et al. (1997)	Second	Sierra Nevada samples from Evans et al. (1997). One sample was removed: ms-2. The concentration is almost two orders of magnitude lower than the other samples in the dataset. The independent age for this site is 15,810 and this sample produces an age of 0.
Phillips et al. (2009)	Second	The full dataset is in Phillips et al. (2009). These samples include a subset of the full dataset that comprises 13 out of 45 samples of Recess Pk and Tioga-4 age. These samples were chosen from the full dataset due to

		internal consistency, composition, clear independent age context, and small snow cover corrections.
Phillips et al. (2001)	Second	Also referred to as 'Legacy Phillips'. Discussed independent age constraints and sample condition and the author decided to remove COM92-18, COM92-20, MK-AT3B-28, MC (four samples), CW-3, CW-7. Included 2 additional samples from one of the same sites in the original study: EI92-52-HI, EI92-54-HI.

Table A5: Summary table explaining all sites, any removed samples, or other issues with the datasets. Second indicates the secondary dataset.

A.1 New England (CRONUS Secondary Site)

The Littleton-Bethlehem moraine site in New England (latitude of 44°N, elevation 400 m) is a bouldery moraine complex. The independent age comes from the associated till sheet that is stratigraphically bracketed by varved sediments. The sediment ages restrict the age of the moraine complex to 13900 ± 250 years before 2010. There was concern about excessive erosion and cover by snow, moss containing mineral matter, and tree cover at this site, so it was not included in the primary calibration dataset. Previous results from other nuclides and more detailed site information are included in Balco et al. (2009).

A.2 Evans et al. (1997) - Sierra Nevada (Non-CRONUS Secondary Site)

Sierra Nevada samples (latitude of 38°N, elevation 3000-3600m) were collected from boulders on moraines for which ages could be assigned based on non-cosmogenic constraints (Phillips, 2015, this issue). Some were close to the sample sites of Nishiizumi et al. (1989). The radiocarbon chronology of the Nishiizumi et al. (1989) samples, which have the same chronology as these samples, was re-evaluated in Phillips (2015, this issue). The chronology is dominated by radiocarbon ages on organic material from sediment cores in glaciated areas. The cosmogenic samples are from valley floor settings that were exposed by final retreat of the Tioga IV glacier, which has been assigned an age of

15810 ± 250 years before 2010. The data from this site have been split into two separate datasets: a low-Cl potassium feldspar dataset and a high-Cl leachate dataset.

A.3 Evans (2001) - Scotland (Non-CRONUS Secondary Site)

Evans (2001) calibrated potassium production rates as well as $P_f(0)$ using data from Scotland (latitude of 58°N, elevation 100-500m). The moraines in Scotland were likely exposed around 11,700±300 years before 2010, which has a regional chronology similar to the CRONUS Scotland site, which is based on varve counting, lake core sedimentology, radiocarbon, pollen analysis, chironomid temperature reconstructions, and correlation with the Greenland Ice Core and models (detailed in Section 3.3). There is a possibility that this site could have been exposed slightly earlier than the CRONUS-Earth site because the moraines could have been exposed at the beginning of the retreat instead of the final deglaciation time of the CRONUS rockfall samples. The quartz samples from Evans (2001) were added to the secondary dataset and used to check the calibration of $P_f(0)$. These samples have production entirely from the low-energy neutron pathway.

A.4 Phillips et al. (2009) (Non-CRONUS Secondary Site)

The Sierra Nevada, California, site is located in western California at approximately 37°N and an elevation range of 2400-3600 m. The original chlorine data was published by Phillips et al. (2009), although the site chronology (Phillips, 2015, this issue) is comprehensively covered elsewhere. Although the samples in this study were not collected as part of the CRONUS-Earth project, they are geographically close to the Baboon Lakes CRONUS-Earth site and supplement the relatively limited dataset from that site. The subset used here was chosen from the larger dataset presented in Phillips et al. (2009) due to the internal consistency, composition, and clear independent age context. Three sets of samples from the Humphreys Basin, the Middle Fork of Bishop Creek, and the Bishop Creek drainage were chosen as secondary calibration samples. These samples were collected from several well-dated moraines representing the Recess Peak terminal position, Tioga IV final deglaciation, and the Tioga IV terminal position, with assigned ages of 13360 ± 250, 15800 ± 500, and 16260 ± 500

years before 2010, respectively (Phillips, 2015, this issue). The independent age constraints were primarily radiocarbon ages from cores in both local and regional lakes, although there is supporting evidence from pollen and shoreline outcrops that is consistent with the radiocarbon chronology. The chlorine samples have a wide range of compositions (from K to Ca), but contain higher chlorine concentrations (16-120 ppm) as well. The whole-rock samples were processed at the New Mexico Tech laboratory.

A.5 Phillips et al. (1996, 2001)(Non-CRONUS Secondary Site)

Initial introduction of the data can be found in Phillips et al. (1996) and the reevaluation of the sites and calibration can be found in Phillips et al. (2001). The calibration contains seven locations (Utah, Idaho, New Mexico, Wales, Ellesmere Island, Northwest Territories, and Arizona) that correspond to a variety of landforms (basalt flows, meteor crater, glacial boulders, glacial bedrock, and beach deposits). The dataset spans a variety of latitudes (19.8-80°N) and elevations (20-2578 m). In general, the dataset uses radiocarbon ages to constrain the site age, but there are some thermoluminescence and K/Ar dates as well, with the assigned calibrated ages ranging from 2.1-49 ka, but most sites are younger than 16 ka.

After talking with the author, some samples were removed based on new information about the landforms obtained since the publication. The main reason for removing samples was doubt about independent age constraints. Removed samples include Meteor Crater (MC), Mauna Kea (MK), and Wales (CW).

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