Long term low latitude and high elevation cosmogenic ^3He production rate inferred from a 107 ka-old lava flow in Northern Chile; 22°S-3400 m a.s.l.

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1 **Abstract**

2

3 Available geological calibration sites used to estimate the rate at which
4 cosmogenic ^3He is produced at the Earth's surface are mostly clustered in
5 medium to high latitudes. Moreover, most of them have exposure histories
6 shorter than tens of thousands of years. This lack of sites prevents a qualitative
7 assessment of available production models used to convert cosmogenic ^3He
8 concentrations into exposure ages and/or denudation rates. It thus limits our
9 ability to take into account the atmospheric, geomagnetic and solar modulation
10 conditions that might have affected the production of cosmogenic nuclides in the
11 past for longer exposure histories and in low latitude regions. We present the
12 cosmogenic ^3He production rate inferred from a new geological calibration site
13 located in northern Chile. Five samples were collected on the surface of the
14 largest and best-preserved lava flow of the San Pedro volcano (21.934°S-
15 68.510°W- 3390 m a.s.l), which displays pristine crease-structure features.
16 $^{40}\text{Ar}/^{39}\text{Ar}$ dating yielded a reliable plateau age of 107 ± 12 ka for the eruption of
17 this lava flow. Eight pyroxene aliquots separated from the surface samples yield
18 a weighted average cosmogenic ^3He concentration of 99.3 ± 1.2 Mat.g^{-1} from
19 which a local cosmogenic ^3He production rate of 928 ± 101 $\text{at.g}^{-1}.\text{yr}^{-1}$ is calculated.
20 The local production rate is then scaled to a sea level high latitude (SLHL)
21 reference position using different combinations of geographic spatialization
22 schemes, atmosphere models and geomagnetic field reconstructions, yielding
23 SLHL production rates between 103 ± 11 and 130 ± 14 $\text{at.g}^{-1}.\text{yr}^{-1}$ consistent with
24 the most recent estimates available from the literature. Finally, we use the same
25 scaling frameworks to re-evaluate the mean global-scale cosmogenic ^3He

26 production rate in olivine and pyroxene minerals at $120 \pm 16 \text{ at.g}^{-1}.\text{yr}^{-1}$ from the
27 compilation of previously published calibration datasets.
28

1. Introduction

Studies relying on dating landforms and quantifying rates of Earth surface processes have greatly benefitted from the recent development of Terrestrial Cosmogenic Nuclides (TCN; see von Blanckenburg and Willenbring, 2014 and references therein for a review). Since the mid-80s (e.g. Craig and Poreda, 1986; Kurz, 1986), cosmogenic helium-3 (^3He) has been successfully used to date geologic features in various environments and over periods of times ranging from hundreds to millions of years (e.g. Kurz et al., 1990; Licciardi et al., 1999; Schäfer et al., 1999; Evenstar et al., 2009). Cosmogenic ^3He combines high production rates ($> 100 \text{ at.g}^{-1}\text{.yr}^{-1}$ at sea level and high latitude) and low analytical detection limits (ca. 10^5 at.g^{-1} in most favorable cases) with the absence of radioactive decay (i.e. ^3He is stable). Cosmogenic ^3He also has the advantage to be produced and quantitatively retained over geological timescales in mafic minerals such as olivines and pyroxenes, which offers an interesting alternative for applying surface exposure dating in volcanic terrains where quartz-bearing lithologies might not be available for ^{10}Be and ^{26}Al measurements.

The rate at which cosmogenic ^3He is produced at the Earth's surface has been estimated between 100 and $140 \text{ at.g}^{-1}\text{.yr}^{-1}$ (see Dunai, 2010 and reference therein) from several geological calibration datasets with independent age assignments (e.g. Ackert et al., 2003; Amidon and Farley, 2011; Blard et al., 2006; 2013; Cerling and Craig, 1994; Dunai and Wijbrans, 2000; Fenton et al., 2013; 2014, Foeken et al., 2012; Goehring et al., 2010; Licciardi et al., 1999; 2006). At a global scale, cosmogenic ^3He production rates at sea level and high latitude

(SLHL) were estimated using the time-dependent adaptation of the Lal (1991)/Stone (2000) scaling scheme ("Lm"; Balco et al., 2008). The available calibration datasets yielded values of 119 ± 9.9 at.g⁻¹.yr⁻¹ for pyroxene (Goehring et al., 2010) and 116 ± 13 (Goehring et al., 2010), 122 ± 15 (Blard et al., 2013) and ca. 117 at.g⁻¹.yr⁻¹ (Borchers et al., 2016) when ³He concentrations of both pyroxene and olivine minerals are considered. Although the global-scale SLHL cosmogenic ³He production rates are consistent within associated uncertainties, Phillips et al. (2016) recently emphasized that the initial datasets used in these compilations are clustered both in space (medium to high latitudes) and time (ca. 5-20 ka). According to these authors, this circumstance prevents reliable integrations of past atmospheric, geomagnetic and solar modulation conditions that might have affected the production of cosmogenic nuclides at the Earth's surface. In particular, cosmogenic ³He calibration data are lacking at latitudes < 25° (Blard et al., 2013), where large temporal variations in the paleo magnetic-field intensity might have significantly affected the production of cosmogenic nuclides (e.g. Dunai and Lifton, 2014). Accordingly, improving the accuracy of time dependent corrections requires more low latitude calibration sites with exposure durations significantly longer than 20 ka.

This article presents a new calibration of the cosmogenic ³He production rate from a site located in the hyperarid region of Northern Chile (3400 m a.s.l., ca. 22°S), over a long exposure time (ca. 107 ka). The aim is to reduce the lack of long exposure calibration sites located at high elevation/low latitude. Five samples were collected for helium analyses on top of an andesitic lava flow. This surface is characterized by pristine crease-structure features, which are common on silicic volcanic flows and previously identified as reliable targets for surface

exposure dating (Anderson et al., 1994). First we document the age of a lava flow of the San Pedro volcano using $^{40}\text{Ar}/^{39}\text{Ar}$ dating techniques. We then report the results of the cosmogenic ^3He measurements from five samples (8 aliquots) from which we infer the local production rate of cosmogenic ^3He , as well as the production normalized to sea level and high latitude (SLHL), both integrated over the age of the lava flow. Finally, we combine our results with previously reported cosmogenic ^3He calibration datasets, based on which we re-evaluate the global ^3He production rate at SLHL.

2. Geological setting and sampling

2.1. Volcanic architecture

The San Pedro volcano (6159 m a.s.l., Figure 1) is situated in the El Loa province of Northern Chile ca. 80 km north east of Calama and 100 km north of San Pedro de Atacama. While the San Pedro volcano is located on the western slope of the High Andean Cordillera and therefore does not belong to the Atacama Desert *stricto sensu*, it is characterized by a hyperarid climate, with annual precipitation < 100 mm/y (Houston and Hartley, 2003). The San Pedro volcano is paired with the San Pablo volcano (6118 m a.s.l.) located 5 km further to the east. It represents one of the highest and best-preserved volcanic edifices of the Central Volcanic Zone (de Silva and Francis, 1991), one of the four active volcanic zones in the Andes (see Hora et al., 2007 and reference therein). Only few studies (Francis et al., 1974; Thorpe et al., 1976 and O’Callaghan and Francis 1986) described and identified the nature and composition of erupted materials and the history of eruption of these volcanoes. The San Pedro volcano represents a

composite andesitic stratovolcano (Figure 2) made of two east-west oriented coalescent cones that have been successively built as the volcanic belt shifted westward (O'Callaghan and Francis, 1986). The cones stand >2 km above an extensive ignimbrite plateau (Figure 1) that has formed since the Late Miocene at ca. 10.4 Ma during the development of the Altiplano-Puna volcanic complex (de Silva, 1989). The San Pedro cones have been built due to the eruption of silicic materials ranging from basaltic andesite to potassium-rich dacite (O'Callaghan and Francis, 1986). Structural mapping and petrologic investigations (Francis et al., 1974; O'Callaghan and Francis, 1986) revealed that the San Pedro Volcano records three evolutionary steps, comprising i) the edification and subsequent collapse of an "old cone", whose summit marks the highest peak of the volcano, ii) the construction, c.a. 5 km to the west, of the "young cone" that has produced several large lava flows and is still marked by fumarolic activity and iii) the formation of the South West Dome and La Poruña scoria cone, representing the last eruptive episode and associated with the emission of two large and well preserved lava flows (Figure 1). Above 4500 m a.s.l., Francis et al. (1974) reported the presence of glacial moraines and cirques on the southern flanks of both the San Pablo and San Pedro volcanoes. Thus, the last eruptions are probably older than the last large glacial advances in the region (i.e. ca. > 15 ka; Blard et al., 2013). In addition, Wörner et al. (2000) reported an exposure age of ca. 103 ka based on the cosmogenic-³He analysis of one sample collected on La Poruña (Figure1), suggesting that the lava flow emitted by the SW Dome might be at least 100 ka old, or even older.

2.2. The SW Dome of the San Pedro volcano

The lava flow emitted by the SW Dome is the largest of the San Pedro volcano. It extends towards the south west for ca. 13 km from ca. 4800 m to 3370 m a.s.l (Figure 1) and covers an area of ca. 20 km² with a volume estimated at 2.8 km³ (O'Callaghan and Francis, 1986). The lava flow is considered the product of a 200-m-thick early pyroxene dacite lava flow that was locally overlain by subsequent hornblende dacite flows (Figure 1). Although most of the surface of the lava flow displays a rough blocky structure typical of low-viscosity Andean andesite/dacite flows, its distal part presents smooth stream-oriented and elongated features (Figure 2). These features correspond to crease structures (Anderson and Fink, 1992) as inferred from the curved and striated walls situated on both sides of a main central fracture (Figure 2). Such crease structures have been identified as typical constructional volcanic features materializing excellent landforms for surface exposure dating with in-situ produced cosmogenic nuclides (Anderson et al., 1994).

2.3. Sampling site

Samples were collected on the top of one crease structure immediately above the main lava flow front. The site is characterized by a perfectly preserved rind crust displaying stream-parallel striations (Table 1, Figures 1 and 2). We collected 5 different samples at a few meters interval in order to capture variations in noble gas concentrations that might be related with differences in the preservation of the flow surface. All sampled surfaces showed no sign of post-eruptive erosion or burial. Seasonal snow cover is unlikely given the hyperaridity of the local climate. Samples were collected with a combination of drill-hammer, sledge-hammer and chisels in order to extract homogeneous <10-cm-thick rock cuboids. Samples'

locations were recorded with a hand-GPS, and both the local surface geometry and its surrounding topography were measured with a sighting compass and inclinometer. These measurements were later used to correct for geomorphic shielding using equations from Dunne et al. (1999).

3. Experimental procedures

The following sections provide a description of the sample preparation and analytical experiments conducted for the determination of the eruption age of the San Pedro volcano lava flow via $^{40}\text{Ar}/^{39}\text{Ar}$ dating and the quantification of the cosmogenic ^3He concentrations of the collected samples.

3.1 Sample preparation

Before proceeding to the preparation of samples for $^{40}\text{Ar}/^{39}\text{Ar}$ dating and helium analyses, the rocks were sawed and 3 separates of each sample were used for estimating the lava density. In addition, the bulk rock composition of major elements was measured on one additional sample separate (Table 1, Table 2). Each sample's average density was determined by weighing several times the sample fragments in air and immersed. Bulk rock composition of collected samples was obtained from mass-spectrometry analyses conducted by Activation Laboratories Ltd. (Canada).

After measuring the sample thickness, materials for $^{40}\text{Ar}/^{39}\text{Ar}$ and helium analyses were crushed via electrodynamic disaggregation using the high-voltage pulsed power selFrag-Lab instrument at the Institute of Geological Sciences of the University of Bern. This crushing equipment was preferred to a usual jaw

crusher because of its efficiency in liberating mono-mineral phenocrysts. It also results in a lower loss of material due to mechanical fining, thereby increasing the recovery of mineral separates. Moreover, it has been shown that the electrodynamic disaggregation of rocks do not affect the noble gas content of liberated minerals (Giese et al., 2009). After crushing, the samples were sieved. The 65-125 microns fraction of the sample SP12-02A was processed in order to extract the potassium-rich groundmass for $^{40}\text{Ar}/^{39}\text{Ar}$ dating purposes. Mafic minerals such as pyroxene, amphibole and associated oxides were first removed using a magnetic separator, after which the remaining material was leached in 10% HNO_3 and rinsed several times with deionized water before small aggregates of clean groundmass were hand-picked under a binocular microscope. About 150 mg were finally used for the $^{49}\text{Ar}/^{39}\text{Ar}$ dating.

Sample preparation for helium analyses consisted of the separation of heavy minerals from the 250-500 microns fraction using bromoform (CHBr_3 , 2.9 g.cm^{-3}) and methylene iodide (CH_2I_2 , 3.3 g.cm^{-3}) heavy liquids. Pure separates of pyroxenes without any adhering lava or solid-inclusions were carefully handpicked under a binocular microscope. Clinopyroxene (augite, prismatic greenish grains) and orthopyroxene (enstatite, orthorhombic brownish grains) were isolated. We used color and shape as criteria for mineral separation during picking. Mineral separates were then sonicated with acetone and rinsed several times with deionized water. About 15 grains of each separate were mounted on epoxy-resin discs in order to measure the equivalent spherical radius of the phenocrysts and to determine the composition in major elements of the mineral separates. This was accomplished using the electron microprobe (JEOL-8200) of the Institute of Geological Sciences in Bern. Laser ablation (LA) ICP-MS analyses

were subsequently conducted at the University of Bern on the two discs of sample SP12-02D (i.e. both the clino- and orthopyroxene separates) in order to measure the concentration of a selection of trace elements, notably U, Th, Sm, Li and Gd (Table 3). These data were later used for correcting the helium analyses for the inherited nucleogenic and radiogenic components.

3.2. $^{40}\text{Ar}/^{39}\text{Ar}$ analytical method

About 150 mg of groundmass from SP12-02A was irradiated for 1 hour (IRR-95) in the OSIRIS nuclear reactor ($\beta 1$ tube, CEA Saclay, France). After irradiation, ca. 40 mg of the sample was transferred into a stainless steel sample holder and then loaded into a differential vacuum Cleartran® window. Argon isotopes were then extracted from the groundmass by step heating using a 25 W CO₂ laser (Synrad®). Ar isotopes were analyzed using a VG5400 mass spectrometer equipped with a single ion counter following the protocol reported in Nomade et al. (2010). Each Ar isotope measurement consisted of 20 cycles of peak jumping over the argon isotopes. Neutron fluence (J) was monitored by co-irradiation of Alder Creek Sanidine (ACs-2, Nomade et al., 2005) placed in the same pit as the sample. J values were determined from analyses of three ACs-2 single grains. Corresponding J value was calculated using an age of 1.194 Ma (Nomade et al., 2005) and the total decay constant reported by Steiger and Jäger (1977). An $^{40}\text{Ar}/^{36}\text{Ar}$ atmospheric ratio of 298.56 was then applied (Lee et al., 2006), consistent with the value reported by Valkiers et al., (2010). Recent revisions of decay and monitor constants suggest values about 0.64 % (Kuiper et al., 2008, 1.201 Ma for ACs-2) and 1.0 % (Renne et al., 2011; 1.2059 Ma for ACs-2) older than used here. However, the implied difference in calibrated age is negligible for

our samples (ca. 1 ka) and therefore remains identical within uncertainty with the age reported here. Procedural blanks were measured every three to four steps depending on the beam size. For a typical 10-minutes static blank, typical backgrounds were about $(2-5) \times 10^{-17}$ and $(4-5) \times 10^{-19}$ moles for ^{40}Ar and ^{36}Ar , respectively. The precision and accuracy of the mass discrimination correction was monitored by daily measurements of air argon at various pressures (Nomade et al., 2010). Nucleogenic production ratios used to correct for reactor produced Ar isotopes from K and Ca could be found in the full data table (Table A1). Data reductions were done using ArArCalc (Koppers, 2002). Plateau age and corresponding uncertainty were calculated using IsoPlot 3.0 (Ludwig, 2003).

3.3. Helium analyses

Helium analyses were carried out at the CRPG noble-gas lab in Nancy, France. ^3He and ^4He isotope concentrations were measured using the Split Flight Tube mass spectrometer designed by GV. The magmatic contribution to the measured ^3He and ^4He concentrations was estimated by a 6 min in-vacuo crushing (Kurz, 1986; Scarsi, 2000) of 97 mg of an aliquot of orthopyroxene of sample SP12-02D. These values were then used for all other samples, assuming that the magmatic contribution is the same in all pyroxenes collected on the San Pedro lava flow. Following Williams et al. (2005) and Blard et al. (2013), we also considered that a potential magmatic contribution is negligible in these micro-phenocrysts.

Between ca. 12 and 28 mg of clino- and orthopyroxene were packed in tin foils and fused in-vacuo for 15 min at 1400°C, within an in-house-built single vacuum furnace (Zimmermann et al., 2012). The extracted gas was purified using a succession of activated-charcoal cold traps, Ti-foam and SAES getters. Cryo-

254 focusing of the gas at various temperatures (12 K and 70 K) thus allowed
255 suppression of neon before helium analysis. Following the purification steps, the
256 total ^3He and ^4He concentrations were analyzed with the SFT mass spectrometer.
257 Re-extraction procedure were conducted on 3 aliquots by heating the furnace to
258 1400°C for an additional 15 min to ensure a complete degasing of the samples.
259 Blank signatures were systematically obtained for these tests. Typical furnace
260 blanks were $(3.3 \pm 1.1) \times 10^4$ and $(1.8 \pm 0.4) \times 10^9$ atoms for ^3He and ^4He ,
261 respectively, which represent <3% of the total ^3He concentrations and ca. 50-
262 80% of the total ^4He measured. We emphasize that such a large contribution by
263 the furnace blank to the total ^4He signal of the samples is actually not related to
264 the quality of the blanks (i.e. blank values obtained during the experiments were
265 consistent with long term blank values) but rather to the very low ^4He content of
266 the samples. The SFT sensitivity was calibrated using a known amount of helium
267 gas standard with a $^3\text{He}/^4\text{He}$ ratio of 20.63 Ra ($1\text{Ra} = 1.384 \times 10^{-6}$ corresponds to
268 the atmospheric ratio) (Matsuda et al., 2002). The reproducibility at the 1-sigma
269 level on the gas standard was 1.5% for ^3He and 0.3% for ^4He over a 2-month-
270 period overlapping with our analyses (1 week).
271 Potential contributions in radiogenic ^4He due to U, Th and Sm decay were
272 estimated following Blard and Farley (2008). The contribution due to
273 nucleogenic ^3He was calculated following Andrews (1985), Andrews and Kay
274 (1982) and using 1) Li concentrations measured in both clino- and
275 orthopyroxene separates of SP12-02D, 2) the compositions in major and trace
276 elements measured in bulk rocks as well as 3) the eruption age of the lava
277 inferred from the $^{40}\text{Ar}/^{39}\text{Ar}$ dating (Table 2, Table 3, Table 4). The concentration
278 of cosmogenic ^3He in our samples ($^3\text{He}_c$) was finally determined using the

equations reported by Kurz (1986) assuming a negligible contribution from radiogenic ^4He (Blard and Farley, 2008):

$$^3\text{He}_c = ^3\text{He}_t - ^3\text{He}_n - ^3\text{He}_m \quad (1)$$

where $^3\text{He}_t$ is the quantity of ^3He measured when fusing the sample, $^3\text{He}_n$ is the nucleogenic component predicted from geochemical analyses of the minerals and bulk rock and $^3\text{He}_m$ the magmatic contribution measured from the in-vacuo crushing of an aliquot of the sample. Note that we do consider here $^3\text{He}_c$ as the product of both spallogenic (induced by high-energy neutrons) and capture reactions (induced by cosmogenic thermal neutrons, CTN). Previous studies reported by e.g. Dunai et al. (2007) and Amidon and Farley (2011) have shown that the contribution of CTN to the cosmogenic ^3He concentration depends on the Li concentration of the analyzed minerals and the thermal neutrons flux that irradiates the considered surface. We emphasize, however, that the CTN contribution to the San Pedro samples is likely negligible due to the limited content in Li (<30 ppm) in both the bulk rocks and pyroxene separates (Table 3) and because of the extreme local climate aridity that strongly reduces the probability of H-rich material (e.g. snow) covering the sampled surfaces, and consequently the thermal neutrons flux (see Dunai et al., 2007). We thus anticipate a CTN contribution lower than 5% to the cosmogenic ^3He concentrations inferred for the San Pedro samples based on observations reported by Amidon et al. (2009) and Blard et al. (2013b).

4. Results

4.1. $^{40}\text{Ar}/^{39}\text{Ar}$ dating of the eruption of the San Pedro Lava flow

The eruption age of the San Pedro volcano lava flow has been estimated from the $^{40}\text{Ar}/^{39}\text{Ar}$ analyses. Results are presented in table 4 and figure 3. The step-heating degassing spectrum yielded a statistically robust plateau age of 107 ± 12 ka (1σ , Mean Square Weighted Deviates, MSWD=0.07) with 100% of the released ^{39}Ar and no rejected heating steps. The age obtained for the total fusion step is consistent with the plateau age at 109 ± 13 ka (Table 4). Alternatively, Ar data were also cast as inverse isochron age. However, this age is poorly constrained at 100 ± 52 ka, mainly because of the low radiogenic content in the sample resulting in a very limited spread on the isochron. We therefore discarded this age when calculating the production rates of cosmogenic ^3He . Instead, we applied the $^{40}\text{Ar}/^{39}\text{Ar}$ plateau age of 107 ± 12 ka for the calibration of the cosmogenic ^3He production rate, as this age yields a good reproducibility at all heating steps. This age is also consistent with the exposure age ca. 103 ka reported by Wörner et al. (2002) for the eruption of La Poruña.

4.2. Helium isotopic results and corrections applied to ^3He .

Helium isotope analyses are reported in Table 5 and cosmogenic ^3He concentrations ($^3\text{He}_c$) calculated for the San Pedro volcano samples are plotted in Figure 4. Note that helium concentrations are reported in “Mat.g $^{-1}$ ” (i.e. 10^6 atoms per gram of pyroxene or ca. 3.7×10^{-14} cc STP/g) in the following sections. Total concentrations released by melting ranged between 89 ± 3 and 97 ± 3 Mat.g $^{-1}$ for ^3He ($^3\text{He}_t$) and between 29 ± 14 and $90 \pm 16 \times 10^3$ Mat.g $^{-1}$ for ^4He ($^4\text{He}_t$). ^3He concentrations cluster around ca. 94 ± 2 Mat.g $^{-1}$, while the ^4He data show a large scatter, which is probably due to the low amount of released gas and the significant contribution of the furnace blank correction (Table 5). In-vacuo

crushing of the aliquot of sample SP12-02D yielded concentrations of
 (0.29±0.09) Mat.g⁻¹ and (17.0±0.6) × 10³ Mat.g⁻¹ for ³He and ⁴He, respectively
 (Table 5). The gas liberated during crushing has thus a ³He/⁴He isotopic ratio of
 12±4 Ra that is higher than the values usually reported for volcanic rocks in the
 Central Andes (Hilton et al., 1993; Blard et al., 2013). This result suggests that
 unquantifiable amounts of cosmogenic ³He might have been released during in-
 vacuo crushing, with the consequence that the magmatic correction applied to
 the samples must be considered as a maximum. However, the impact of this
 magmatic correction is most likely negligible because the ³He released during
 crushing (³He_m) only represents ca. 0.3% of the total ³He released during melting
 (³He_t, Table 5). Note that the magmatic ³He correction applied to all the samples
 was realized using the ³He concentration measured from the prolonged in-vacuo
 crushing experiment of the sample SP12-02D. This correction systematically
 represented less than 0.4% of the total ³He extracted by fusion in all samples and
 thus does not induce a significant source of uncertainty.

Regarding ⁴He, the concentration we measured during the crushing experiment
 (17 ± 0.6) × 10³ Mat.g⁻¹ represents ca. 20% of ⁴He_t determined in the
 orthopyroxene aliquot of SP12-02D (77±23) × 10³ Mat.g⁻¹ (Table 5). If we
 consider that this ⁴He concentration measured during crushing is magmatic and
 representative for all samples, then 40 to 80% of ⁴He_t is most likely radiogenic in
 other samples. Using the radiogenic production rates estimated from the
 minerals composition (P_{4r}: ca. 1.1±0.1 Mat.g⁻¹.yr⁻¹, Table 3), the calculated (U-Th-
 Sm)/⁴He ages vary from 12 to 54 ka, i.e. significantly younger than the ⁴⁰Ar/³⁹Ar
 age of 107±12 ka. Assuming no magmatic ⁴He correction, these (U-Th-Sm)/⁴He
 ages range from 26 to 76 ka. Such a difference with the ⁴⁰Ar/³⁹Ar age may

indicate that the U, Th and Sm decay chains are out of secular equilibrium (Farley et al., 2002). In any case, given the large uncertainties related to the measurement of ^4He concentrations and to the production rate, the (U-Th-Sm)/ ^4He approach cannot be used to thoroughly constrain the age of the flow eruption. We will therefore rely on the $^{40}\text{Ar}/^{39}\text{Ar}$ age only to calibrate the cosmogenic ^3He production rate at the San Pedro volcano.

The nucleogenic ^3He ($^3\text{He}_n$) production rates range between ca. 0.1 and 0.2 at.g $^{-1}$.yr $^{-1}$ (Table 5). The Li concentrations used for this calculation are 20 ppm Li for clinopyroxene and 15 ppm Li for orthopyroxene (from LA ICP-MS analyses of sample SP12-02D, Table 3). Although these values have been used for all 5 samples, we emphasize that this simplification does not induce significant uncertainties on the nucleogenic ^3He correction as $^3\text{He}_n$ represents indeed less than 0.03% of $^3\text{He}_t$ (Table 5).

4.3. Calculation of cosmogenic ^3He concentrations

Cosmogenic ^3He ($^3\text{He}_c$) reported in Table 5 are corrected for inherited magmatic and nucleogenic components. They range between 89 ± 3 and 96 ± 3 Mat.g $^{-1}$. Subsequent corrections for thickness, geomorphic shielding and normalization to a geographic position of 21.934°S-68.510°W-3390 m a.s.l. (i.e. total relative correction of ca. 4.5-8.7 %, see Table 1) yield surface cosmogenic ^3He concentrations ($^3\text{He}_{c(S)}$) that range from 93 ± 3 to 102 ± 3 Mat.g $^{-1}$, with an arithmetic mean of 99 ± 3 Mat.g $^{-1}$.

The $^3\text{He}_{c(S)}$ concentrations obtained for clinopyroxene and orthopyroxene separates are similar within uncertainties. This is consistent with results from elemental production models (e.g. Masarik and Reedy, 1996) that do not predict

any significant differences in production rate between these two pyroxene phases. Weighted average values can therefore be calculated from clino- and orthopyroxene for the same samples. The narrow distribution displayed by the data thus suggests that all sampled surfaces have experienced the same exposure history. This indicates that the surface of the San Pedro lava flow has not been affected by post-eruptive erosion or burial, consistent with our observations in the field. This also implies that the San Pedro site is suitable for calibrating the production rate of cosmogenic ^3He . In summary, the five samples finally yield a weighted average of $99.3 \pm 1.2 \text{ Mat}_{(^3\text{He})} \cdot \text{g}_{(\text{pyroxene})}^{-1}$ with a relative standard deviation of ca. 1% and a MSWD of 1.2 (Figure 4).

5. Discussion

5.1. Estimations of cosmogenic ^3He production rates

The production rate of cosmogenic ^3He can be estimated locally, at the geographic position of the calibration site, and/or scaled to a sea level high latitude (SLHL) reference position. Locally, the cosmogenic ^3He production rate is obtained by dividing the average cosmogenic ^3He concentration obtained for the calibration site by the age of the considered landform deduced from an independent dating technique (e.g. ^{14}C , K/Ar, $^{40}\text{Ar}/^{39}\text{Ar}$). Note that individual concentrations are first normalized to the surface, an open-sky topography and the mean geographic coordinates (latitude, longitude and elevation) in order to identify potential outliers due to erosion or inheritance (see e.g. Martin et al., 2015, and reference therein). Scaling the cosmogenic ^3He production rate to SLHL (referred to as SLHL P3 in following sections) is, however, not

straightforward. The scaling procedure indeed requires the combination of a geographic spatialization model with an atmosphere model and the reconstruction of the past geomagnetic field conditions (see Balco et al., 2008; Lifton et al., 2014). Here we followed a calculation procedure based on a least-square minimization technique (see Martin et al., 2015, for details) and the scaling frameworks presented bellow. SLHL P3 estimates were inferred from the San Pedro samples using the time-independent Lal/Stone scaling model (Lal, 1991; Stone, 2000), noted “St”, as well as two time-dependent schemes: “Lm”, the adaptation of the Lal/Stone model (Balco et al., 2008) and “LSD”, recently developed by Lifton et al. (2014). SLHL P3 estimates were thus obtained by dividing the local production rate by a scaling factor representative for a given combination of scaling, atmosphere and geomagnetic reconstruction models. Regarding scaling schemes, we favored the “Lm” and “LSD” time-dependent models. Indeed, recent studies (Lifton et al., 2014; Borchers et al., 2016; Phillips et al., 2016) have demonstrated that these scaling schemes yield more consistent and reliable spatial and temporal estimates of cosmogenic nuclide production rates than former neutron-monitor based models (Desilets and Zreda, 2003; Desilets et al., 2006; Dunai, 2001; Lifton et al., 2005). These “Lm” and “LSD” models thus represent the most widely used and recently available scaling schemes, respectively (see e.g. Lifton et al., 2014 and Martin et al., 2015). We then calculated SLHL P3s for “St”, “Lm” and “LSD” using the Standard atmosphere model (National Oceanic and Atmospheric Administration, 1976) and the ERA-40 reanalysis dataset (Uppala et al., 2005), which allows accounting for the regional variations in height-pressure relationship. Finally, we inferred SLHL P3s for the time-dependent “Lm” and “LSD” scaling schemes with three

paleomagnetic-field reconstruction models (see details regarding the geomagnetic field reconstructions in Martin et al., 2015). The first two geomagnetic models rely on Virtual Dipole Moment values based on 1) Muscheler et al. (2005) for 0-62 ka BP and SINT-2000 (Valet et al., 2005) beyond and 2) GLOPIS-75 (Laj et al., 2004) for 0-75 ka BP and SINT-2000 beyond. The third model is based on the composite reconstruction of temporal variations in geomagnetic and solar intensity modulations implemented in the LSD model (Lifton et al., 2014).

5.2. Calibration of $^3\text{He}_c$ production rates at 21.934°S-68.510°W-3390 m a.s.l.

The local cosmogenic ^3He production rate, estimated for the San Pedro calibration site at a geographic position of 21.934°S-68.510°W and 3390 m a.s.l., is obtained by dividing the weighted average $^3\text{He}_{c(s)}$ with the eruption age of the San Pedro lava flow obtained from the $^{40}\text{Ar}/^{39}\text{Ar}$ dating. This approach yields a local production rate of $928 \pm 11 \text{ at.g}^{-1}.\text{yr}^{-1}$, and $928 \pm 101 \text{ at.g}^{-1}.\text{yr}^{-1}$ when propagating both the cosmogenic ^3He and $^{40}\text{Ar}/^{39}\text{Ar}$ uncertainties respectively, at the 1-sigma level (Table 5). This rate combines all production pathways of cosmogenic ^3He including spallation, thermal neutron capture and muogenic production. We note however, that the contribution of cosmogenic thermal neutrons to the production of cosmogenic ^3He at the San Pedro calibration site is likely lower than 5% and therefore negligible, based on the predictions obtained with the model reported by Amidon and Farley (2011) for pyroxenes with 15-20 ppm apparent Li and a He-closure age of ca. 100 ka.

The local cosmogenic ^3He production rate is then scaled to a sea level and high latitude (SLHL) reference position. SLHL P3s inferred from the San Pedro

samples are presented in Table 6. SLHL P3s obtained from the time-independent
 “St” scheme with the Standard and ERA-40 atmosphere models are 132 ± 14 and
 143 ± 16 $\text{at.g}^{-1}.\text{yr}^{-1}$, respectively. SLHL P3s estimated with the time-dependent
 “Lm” and “LSD” schemes, with various combinations of atmosphere and
 geomagnetic models, vary from 103 ± 11 to 130 ± 14 $\text{at.g}^{-1}.\text{yr}^{-1}$ (Table 6). In
 general, the inferred values agree well with the SLHL P3 reported from global-
 scale compilations by Goehring et al. (2010) and more recently by Blard et al.
 (2013a). Moreover, SLHL P3s that we estimated using the time-dependent
 adaptation of the Lal/Stone scaling scheme (“Lm”) with the Muscheler-SINT2000
 geomagnetic reconstruction and considering both the Standard and ERA-40
 atmosphere models are 120 ± 13 and 130 ± 14 $\text{at.g}^{-1}.\text{yr}^{-1}$, respectively.
 Interestingly, these SLHL P3 estimates fit perfectly with the values reported by
 Blard et al. (2013a) from a ca. 15-ka-old calibration site located at 3800 m a.s.l.
 on the Bolivian Altiplano (Figure 1).
 Note that all applied time-dependent scaling frameworks predict that at the San
 Pedro volcano calibration site, the production of cosmogenic nuclides, including
 cosmogenic ^3He , has been generally higher during the last 100 to 150 ka than at
 present (Figure 5). Such a situation can be explained by the persistence of a
 weaker geomagnetic field intensity that has induced lower cutoff rigidities,
 thereby allowing more cosmic-ray particles to enter the Earth’s magnetic field
 (see Dunai and Lifton, 2015 and references therein). This effect is particularly
 amplified in high tropical areas integrating >50-ka exposure histories (e.g. Balco
 et al., 2008), since it yields relative corrections between 10 and 20% of the
 modern rate (Figure 5). It thus explains why the San Pedro calibration site yields
 time-independent “St” scaling factors that are higher than those inferred from

time-dependent schemes (“Lm” and “LSD”). Figure 5 and Table 6 illustrate that geomagnetic field reconstructions relying on the Muscheler-SINT2000 and “LSD” frameworks systematically yield the highest and lowest SLHL P3s, respectively, independently of the scaling scheme and atmosphere model considered. The geomagnetic field and solar modulation reconstructions implemented in the “LSD” frameworks predict time-integrated scaling factors that are up to ca. 10% higher than with the Muscheler-SINT2000 for an exposure length > 60 ka.

5.3. Re-evaluation of the global SLHL cosmogenic ^3He production rate

Cosmogenic ^3He production rates have rarely been calibrated to sea level high latitude positions using a common calculation framework that included similar scaling schemes, atmosphere model and geomagnetic field reconstructions. Here we re-evaluate the global SLHL P3 by compiling and processing available calibration datasets for both olivine and pyroxene within a common calculation framework that combines the scaling schemes, atmosphere and geomagnetic field models presented above (see section 3.4). Twenty different calibration sites and ca. 150 cosmogenic ^3He analyses have been compiled from the literature and combined with the San Pedro samples (see Table A2). This new compilation includes the studies of Ackert et al. (2003), Amidon and Farley (2011), Blard et al. (2006, 2013a), Cerling and Craig (1994); Dunai and Wijbrans (2000), Fenton et al. (2013, 2014), Foeken et al. (2012), Goehring et al. (2010) and Licciardi et al. (1999, 2006). We do note that we only considered dataset originating from absolute calibration studies and therefore excluded those from cross calibrations (e.g. Amidon et al, 2009). Some of the studies included in this compilation did not address the impact of radiogenic ^4He on the measured cosmogenic ^3He

504 concentrations. In several of these previous studies indeed, the authors
 505 overestimated the amount of ^4He that was derived from trapped magmatic gases
 506 and thus over-corrected for magmatic ^3He , thereby resulting in an
 507 underestimation of cosmogenic ^3He concentrations. To tackle this issue, the
 508 compiled data were first corrected for this radiogenic contribution, either using
 509 the reported U-Th-Sm concentrations in the phenocrysts or considering the
 510 global mean of these elements as proposed by Blard and Farley (2008, see Table
 511 A2). The resulting corrections slightly increase the cosmogenic ^3He
 512 concentrations by 4% on average, as illustrated in the inset of Figure 6A.
 513 Subsequently, the cosmogenic ^3He concentrations available for each calibration
 514 site were 1) normalized to the surface, an open-sky topography and the sites'
 515 mean geographic coordinates, 2) used to calculate a weighted average
 516 concentration and 3) inverted to SLHL P3s, following the procedure outlined in
 517 section 3.4. SLHL P3s inferred from all possible combinations in scaling,
 518 atmosphere and geomagnetic field models cover the range $76\pm4 - 186\pm12 \text{ at.g}^{-1}\text{.yr}^{-1}$
 519 (see Table A2). When considering each scaling framework separately, the
 520 values yield arithmetic means that range between 115 ± 19 and $122\pm20 \text{ at.g}^{-1}\text{.yr}^{-1}$,
 521 therefore identical within uncertainty (1σ -level). These re-evaluated global SLHL
 522 P3s are consistent with values reported from previous compilations that also
 523 considered olivine and pyroxene minerals together (e.g. Goehring et al., 2010;
 524 Blard et al., 2013a; Borchers et al., 2016). We emphasize, however, that SLHL P3s
 525 estimated from the combination of the “Lm” scaling scheme with the ERA-40
 526 atmosphere model and the geomagnetic field reconstruction based on Muscheler
 527 (2004) and SINT2000 (Valet et al., 2004) yield the least scatter in the
 528 distribution, with a 14% relative standard deviation and an arithmetic mean of

120±16 at.g⁻¹.yr⁻¹. Figure 6 represents the distribution of SLHL P3s inferred from this latter scaling framework. It does not show any correlation with elevation nor with latitude. This confirms previous observations reported by Goehring et al. (2010) and Blard et al. (2013a) that there is no anomalous production of cosmogenic ³He at high elevations, at least in pyroxene and olivine. It is, however, important to emphasize that the estimated Mean Square Weighted Deviation is significantly higher than 1 (MSWD = 10.4; Figure 6), indicating that the observed variability in the data is larger than a statistical dispersion that would result from analytical uncertainties only. In other words, the global SLHL P3 compilation unlikely represents a single population. Note that while Figure 6 only represents SLHL P3s estimated from the combination of the “Lm” scaling scheme with the ERA-40 atmosphere model and Muscheler-SINT2000 based geomagnetic field reconstruction, all other scaling frameworks present a similar picture. Part of this over-dispersion might result from calibration sites older than ca. 120 ka that systematically yield SLHL P3s lower than ca. 110 at.g⁻¹.yr⁻¹ when considering the “Lm” scaling scheme (Figure 6). This inference, however, should be tuned down when considering the “LSD” scaling scheme, as P3s estimates do not show such temporal pattern. In any cases and for all considered scaling frameworks, the variability in SLHL P3s seems independent from the exposure age for calibration sites younger than 110 ka (Figure 6). The observed variability in SLHL P3s might result from unrecognized erosion, variations in atmospheric pressure, inaccurate correction of the time-dependent production rate variability, and/or unaccounted excess in Li concentrations of crustally contaminated magmas (e.g. Amidon et al., 2011; Dunai and Wijbrans, 2000; Dunai et al., 2007; Foeken et al., 2012; Staiger et al., 2007). Excluding calibration

sites older than 120 ka from the global compilation leads to slightly higher average cosmogenic ^3He production rates by ca. 2-4% and reduces the standard deviations from 14-17% to ca. 11-17%, depending on the considered scaling framework (see Table A2), but do not fundamentally change the overall picture. In summary, SLHL P3s inferred from the combination of the “Lm” scaling scheme with the ERA-40 atmosphere model and Muscheler-SINT2000 based geomagnetic field reconstruction yield the best constrained mean global-scale cosmogenic ^3He production rate. This scaling framework therefore represents a qualitative combination of currently available geographic spatialization schemes, atmosphere as well as geomagnetic field reconstruction models to estimate exposure ages from cosmogenic ^3He concentrations. Similar statements were also reported by Martin et al. (2015) regarding the production of in-situ ^{10}Be in the high tropical Andes.

6. Conclusion

We report a new geological calibration site located in northern Chile (21.934°S-68.510°W) at 3390 m a.s.l. that we used to infer the production rate of cosmogenic ^3He in pyroxenes. $^{40}\text{Ar}/^{39}\text{Ar}$ dating gives a consistent plateau age of 107 ± 12 ka. Eight mineral separates of clino- and orthopyroxene from five different surface samples yield clustered ^3He concentrations, for both pyroxene phases. Magmatic and nucleogenic ^3He corrections are negligible ($< 1\%$) in this lava flow. The weighted average cosmogenic ^3He concentration is 99.3 ± 1.2 Mat.g $^{-1}$, which represents the highest cosmogenic ^3He concentrations ever measured so far in natural calibration samples.

We then used this data to constrain a local cosmogenic ^3He production rate of $928 \pm 101 \text{ at.g}^{-1}\text{.yr}^{-1}$ at 21.934°S - 68.510°W and 3390 m a.s.l. After scaling to sea level and high latitudes (SLHL), cosmogenic ^3He production rate estimates vary from 103 ± 11 to $130 \pm 14 \text{ at.g}^{-1}\text{.yr}^{-1}$, which depends on different combinations of scaling schemes, atmosphere models and geomagnetic field reconstructions. Finally we combined the SLHL P3s inferred from the San Pedro lava flow samples with cosmogenic ^3He production rates (in both olivine and pyroxene) inferred from various calibration sites and re-evaluated the mean global SLHL P3. This compilation, where cosmogenic ^3He production rates were scaled to SLHL with two scaling schemes, two atmosphere models and three different geomagnetic field reconstructions, yield arithmetic means for SLHL P3s that vary between 115 ± 19 and $122 \pm 20 \text{ at.g}^{-1}\text{.yr}^{-1}$. These values are identical within uncertainty and consistent with values previously reported from similar compilation studies (e.g. Goehring et al., 2010; Blard et al., 2013a; Borchers et al., 2016). This new compilation thus illustrates that a calculation framework including the (1) time-dependent adaptation of the Lal (1991) / Stone (2000) scheme, (2) ERA-40 atmosphere model (Uppala et al., 2005), and a (3) geomagnetic field reconstruction relying on the Virtual Dipole Moment values based on Muscheler et al. (2004) and SINT-2000 (Valet et al., 2005) currently yields the best constrained mean global-scale cosmogenic ^3He production rate. Our observations also appear perfectly in line with the recent findings reported by Lifton (2016). We thus suggest that the update of the scaling framework presented here, together with the inferred mean global SLHL P3 of $120 \pm 16 \text{ at.g}^{-1}\text{.yr}^{-1}$, should be favored when calculating exposure ages from cosmogenic ^3He concentrations.

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Table 1. Sample characteristics

Sample name	Latitude	Longitude	Elevation	Atmospheric pressure	Altitude factor ^b	Sample thickness	Density	Thickness correction ^c	Geomorphic shielding ^d	Total correction ^e
	(°)	(°)	(m)	(hPa) ^a		(cm)	(g.cm ⁻³)			(%)
SP12-02A	-21.93434	-68.50980	3389	679.55	0.999	6	2.44	0.956	0.996	4.9
SP12-02C	-21.93431	-68.50974	3394	679.13	1.002	6	2.41	0.956	0.995	4.6
SP12-02D	-21.93440	-68.50966	3393	679.22	1.002	6	2.43	0.956	0.998	4.5
SP12-02E	-21.93435	-68.50983	3391	679.38	1.001	7	2.22	0.953	0.998	4.9
SP12-02F	-21.93437	-68.51005	3384	679.97	0.997	7.5	2.21	0.950	0.964	8.7

^a Local atmospheric pressure was calculated using the ICAO standard atmosphere equation with sea level temperature and pressure derived from the ERA-40 reanalysis dataset (Uppala et al., 2005)

^b Correction factors used to scale cosmogenic ³He concentrations to 21.934°S-68.510°W, 3390m (679.468 hPa). Values are calculated as the ratio between the scaling factor obtained using the Lal (1991) / Stone (2000) scheme with the ERA-40 atmosphere model for the sampling position and at 21.934°S-68.510°W, 3390 m a.s.l.

^c Thickness corrections are calculated using an attenuation length of 160 g.cm⁻² and the sample densities measured.

^d Total geomorphic shielding, including self-shielding by the sampled surface and the topography around the sampling site, are calculated according to Dunne et al. (1999) and an algorithm provided by G. Balco (<http://hess.ess.washington.edu/>).

^e Calculated as the product of altitude, thickness and geomorphic shielding factors.

Table 2. Major element composition of lava and pyroxene phenocrysts

Sample name	Type	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O	TiO ₂	P ₂ O ₅	LOI	Total
		(wt %)	(wt %)	(wt %)	(wt %)	(wt %)	(wt %)	(wt %)	(wt %)	(wt %)	(wt %)	(wt %)	(%)
SP12-02A	Bulk rock	63.3	15.6	5.5	0.08	2.4	4.1	4.0	3.3	0.8	0.2	1.2	100
SP12-02A	Clinopyroxene	52.1	2.4	9.8	-	14.7	19.6	0.5	0.0	0.5	-	-	100
SP12-02A	Orthopyroxene	54.0	0.9	20.0	-	22.9	1.2	0.0	0.0	0.4	-	-	100
SP12-02C	Bulk rock	65.1	14.7	5.4	0.08	2.2	3.9	3.7	3.1	0.8	0.2	1.5	101
SP12-02C	Clinopyroxene	52.7	2.2	8.9	-	14.9	20.4	0.4	0.0	0.4	-	-	100
SP12-02D	Bulk rock	62.8	15.2	5.5	0.08	2.3	4.1	3.7	3.0	0.8	0.2	1.7	99
SP12-02D	Clinopyroxene	52.0	2.4	10.0	-	14.8	19.3	0.5	0.0	0.6	-	-	100
SP12-02D	Orthopyroxene	53.6	1.1	20.5	-	23.1	1.3	0.0	0.0	0.2	-	-	100
SP12-02E	Bulk rock	61.4	15.2	5.4	0.07	2.2	4.6	3.9	3.0	0.8	0.2	2.0	99
SP12-02E	Clinopyroxene	53.0	1.9	8.8	-	14.8	20.3	0.4	0.0	0.4	-	-	100
SP12-02F	Bulk rock	63.8	14.8	5.2	0.08	2.1	4.2	3.7	2.9	0.7	0.2	1.7	99
SP12-02F	Clinopyroxene	52.7	2.0	9.9	-	14.3	20.0	0.5	0.0	0.4	-	-	100
SP12-02F	Orthopyroxene	54.1	0.8	19.4	-	23.4	1.1	0.0	0.0	0.3	-	-	99

Table 3. Trace element composition of lava and pyroxene phenocrysts

Sample name	Type	Li	B	Cr	Co	Ni	Gd	U	Th	Sm
		(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)
SP12-02A	Bulk rock	18	41	90	12	20	4.7	2.6	9.9	5.6
SP12-02A	Clinopyroxene	-	-	820	-	392	-	-	-	-
SP12-02A	Orthopyroxene	-	-	184	-	174	-	-	-	-
SP12-02C	Bulk rock	16	27	70	11	20	4.0	2.5	8.5	5.0
SP12-02C	Clinopyroxene	-	-	802	-	301	-	-	-	-
SP12-02D	Bulk rock	21	33	90	12	20	4.3	2.5	9.4	5.3
SP12-02D	Clinopyroxene	24	0.7	946	-	725	7.4	<0.01	0.06	7.2
SP12-02D	Orthopyroxene	16	0.6	245	-	185	0.7	<0.01	<0.01	0.4
SP12-02E	Bulk rock	20	32	80	11	20	4.1	2.4	9.1	5.3
SP12-02E	Clinopyroxene	-	-	410	-	384	-	-	-	-
SP12-02F	Bulk rock	18	42	70	11	20	4.2	2.2	8.3	5.2
SP12-02F	Clinopyroxene	-	-	366	-	142	-	-	-	-
SP12-02F	Orthopyroxene	-	-	109	-	171	-	-	-	-

Major and trace elements in lava (bulk rock) were measured by ICP-OES and ICP-MS, respectively, in aliquots of ca. 1 g.

Major element concentrations in phenocrysts represent mean values that were determined by electron-microprobe on 10-15 grains per sample. Absolute standard deviations associated with mean values are <1% in mass (1σ).

MnO and P2O5 analyses in phenocrysts were substituted by NiO and Cr2O3 (values are shown as traces and represent <0.2 % in mass of the total pyroxene composition)

Trace element concentrations in phenocrysts represent mean values that were determined by Laser ICP-MS on ca. 15 grains per samples. Relative standard deviations (1σ) represent ca. <15% (Li), 40% (B, Gd and Sm) and >50% (U and Th).

Table 4. Summary of the SP12-02A ^{40}Ar - ^{39}Ar step heating experiment.

Incremental heating	$^{40}\text{Ar}/^{39}\text{Ar}$	$^{37}\text{Ar}/^{39}\text{Ar}$	$^{36}\text{Ar}/^{39}\text{Ar}$	^{40}Ar (%)	K/Ca	^{39}Ar (%)	Age $\pm 1\sigma$ (ka)
#1	0.1547	0.1727	0.0381	1.36	2.49	9.87	98.9 $\pm$ 24.9
#2	0.1708	0.1384	0.0587	0.98	3.11	19.14	109.2 $\pm$ 39.8
#3	0.1476	0.2527	0.0507	0.98	1.70	7.50	94.3 $\pm$ 35.6
#4	0.1805	0.1416	0.0407	1.48	3.04	22.30	115.4 $\pm$ 23.8
#5	0.1873	0.3236	0.0529	1.18	1.33	10.31	119.8 $\pm$ 40.1
#6	0.1765	0.2447	0.0580	1.02	1.76	14.77	112.8 $\pm$ 37.5
#7	0.1752	0.4887	0.0904	0.65	0.88	5.70	112.0 $\pm$ 73.5
#8	0.1587	0.3469	0.0509	1.04	1.24	10.40	101.5 $\pm$ 30.3
Total fusion	0.1712	0.2275	0.1042		1.89		109.4 $\pm$ 13.2
Plateau	0.1674				1.40		107.0 $\pm$ 11.6

Table 5. Helium analyses

Sample Name	Mineral	Mass (mg)	$^3\text{He}_t$ (Mat.g ⁻¹) ^a	$^4\text{He}_t$ (10 ³ Mat.g ⁻¹) ^a	$P3_n$ (at.g ⁻¹ .yr ⁻¹) ^b	$^3\text{He}_n$ (Mat.g ⁻¹) ^c	$P4_r$ (Mat.g ⁻¹ .yr ⁻¹) ^d	$^3\text{He}_c$ (Mat.g ⁻¹) ^e	$^3\text{He}_{c(S)}$ (Mat.g ⁻¹)	$P^3\text{He}_c$ (at.g ⁻¹ .yr ⁻¹) ^f
SP12-02A	Clinop.	22.9	96.0±2.6	89.9±15.7	0.2	0.02	1.19	95.7±2.6	100.7±2.7	941±25 (105)
SP12-02A	Orthop.	12.2	96.6±3.3	29.9±29.5	0.1	0.01	1.16	96.3±3.3	101.3±3.4	946±32 (108)
<i>SP12-02A</i>	<i>Average</i>	-	-	-	-	-	-	<i>95.9±2.0</i>	<i>100.9±2.1</i>	<i>943±20 (104)</i>
SP12-02C	Clinop.	27.8	91.9±2.4	71.6±13.0	0.2	0.02	1.12	91.6±2.4	96.1±2.5	898±23 (100)
SP12-02D	Clinop.	18.1	95.7±2.8	35.6±20.6	0.2	0.02	1.17	95.4±2.8	99.9±2.9	934±27 (105)
SP12-02D	Orthop.	16.4	89.2±2.8	76.9±22.8	0.1	0.01	1.11	88.9±2.8	93.0±2.9	870±27 (98)
<i>SP12-02D</i>	<i>Average</i>	-	-	-	-	-	-	<i>92.2±3.3</i>	<i>96.5±3.4</i>	<i>902±32 (103)</i>
SP12-02E	Clinop.	25.9	93.5±2.4	29.2±14.4	0.2	0.02	1.13	93.2±2.5	98.0±2.6	916±24 (102)
SP12-02F	Clinop.	25.9	93.0±2.3	29.2±14.4	0.1	0.02	1.04	92.7±2.3	101.5±2.6	949±24 (106)
SP12-02F	Orthop.	23.4	92.6±2.4	37.1±15.4	0.1	0.01	0.98	92.3±2.4	101.1±2.6	945±25 (106)
<i>SP12-02F</i>	<i>Average</i>	-	-	-	-	-	-	<i>92.5±1.7</i>	<i>101.3±1.8</i>	<i>947±17 (104)</i>

^a ^3He and ^4He concentrations measured by fusing the sample and corrected for blanks. Typical ^3He and ^4He blanks during the measurements were $(3.3\pm1.1) \times 10^4$ atoms and $(1.8\pm0.4) \times 10^9$ atoms, respectively, or <3% of ^3He and ca. 50-80% of ^4He measured in the samples. Associated errors (1σ) include sample and blank counting statistics as well as the one-sigma reproducibility on standard analyses over a 2 months period (1.5% for ^3He and 0.3% for ^4He).

^b Nucleogenic ^3He production rates were calculated according to Andrews (1985) and Andrews and Kay (1982) using Li concentration measured in pyroxene aliquots and major and trace element compositions of lava (Table 2).

^c Total nucleogenic ^3He calculated as the product of $P3_n$ and the age of the lava based on the Ar/Ar dating.

^d Radiogenic ^4He production rate were predicted from equations described in Farley et al. (2006) and Blard and Farley (2008). Calculations are based on U, Th and Sm concentrations measured both in bulk rocks and clinopyroxene and orthopyroxene aliquots of sample SP12-02D (see Table 3) together with a measured equivalent spherical radius of 215 μm for pyroxene grains and an estimated 20- μm alpha-nuclei stopping distance.

^e Corrected for magmatic ($^3\text{He}_m$) and nucleogenic ($^3\text{He}_n$) contributions to $^3\text{He}_T$. The magmatic contribution was inferred from a 6 min in-vacuo crushing of an aliquot of sample SP12-02D that yielded concentrations of (0.29 ± 0.09) Mat.g⁻¹ and $(17.0\pm0.6) \times 10^3$ Mat.g⁻¹ for ^3He

and ^4He , respectively, which represented a $^3\text{He}/^4\text{He}$ isotopic ratio of 12 ± 4 Ra. Reported errors (1σ) integrate ca. 3% uncertainty on $^3\text{He}_t$ measurements and 10% and 31% uncertainties on nucleogenic and magmatic contribution estimates, respectively.

^f Local cosmogenic ^3He production rates estimated from $^3\text{He}_c$ concentrations scaled to the surface, an open-sky topography and 21.934°S - 68.510°W and 3390 m a.s.l. and the age of the lava based on the $^{40}\text{Ar}/^{39}\text{Ar}$ dating. Bracketed values correspond to external errors (1σ) and include the ca. 3% uncertainty on $^3\text{He}_c$ concentrations and 11% on the $^{40}\text{Ar}/^{39}\text{Ar}$ inferred age of the lava.

Table 6. SLHL ^3He production rates from the San Pedro calibration site

Scaling scheme ^a	Atmosphere model ^b	Geomagnetic reconstruction ^c	SLHL P3 (at.g ⁻¹ .yr ⁻¹) ^d
“St”	Standard	-	132±14
	ERA-40 (1)	-	143±16
“Lm”	Standard	Musheler-SINT2000	120±13
		GLOPIS75-SINT2000	118±13
		LSD	109±12
	ERA-40	Musheler-SINT2000 (2)	130±14
		GLOPIS75-SINT2000 (3)	128±14
		LSD (4)	118±14
“LSD”	Standard	Musheler-SINT2000	116±13
		GLOPIS75-SINT2000	113±12
		LSD	103±11
	ERA-40	Musheler-SINT2000 (5)	127±14
		GLOPIS75-SINT2000 (6)	124±14
		LSD (7)	112±12

^a Cosmogenic ^3He production rates are scaled to the reference sea level high latitude (SLHL) position using the calculation procedures and employed the scaling frameworks reported by Martin et al. (2015). “St” refers to Lal (1991) / Stone (2000), “Lm” to the time-dependent adaptation of the Lal/Stone scheme (see e.g. Balco et al., 2008) and “LSD” to Lifton et al. (2014).

^b The Standard atmosphere model is based on the National Oceanic and Atmospheric Administration (1976) and the ERA-40 reanalysis dataset on Uppala et al., (2005).

^c Geomagnetic models rely on Virtual Dipole Moment values based on Muscheler et al. (2005), GLOPIS-75 (Laj et al., 2004) and SINT-2000 (Valet et al., 2005). Numbers in brackets refer to scaling factor curves as labeled in Figure 5.

^d SLHL P3 estimates are obtained by dividing the local production rate by a scaling factor inferred for a given combination of scaling, atmosphere and geomagnetic reconstruction models (see text for details). Reported errors correspond to external (total) uncertainties at 1 σ -level and comprise a ca. 1% uncertainty associated with the weighted average cosmogenic ^3He concentration and ca. 11% from the $^{40}\text{Ar}/^{39}\text{Ar}$ dating.

Figure captions

Figure 1. Geological sketch of the San Pedro volcano showing the location of the sampling site. The map is modified after Francis et al. (1974) and O'Callaghan and Francis (1986). Hillshade model and 100-m-interval contours are based on the ASTER GDEM 30-m-resolution digital topography. False easting and northing kilometer coordinates are based on UTM zone 19S projection. Inset shows the location of the San Pedro volcano in the Central Andes, the Atacama Desert as well as the Cerro Tunupa calibration site from Blard et al. (2013a).

Figure 2. Photograph collection of the sampling site. (A) General overview of the San Pedro volcano photographed from the SW lava flow at ca. 3600 m a.s.l. toward the NE. (B) Satellite image (Google Earth) showing the situation of samples (see red star in Figure 1). Note the lobate morphology of the lava front and the stream-oriented elongated features that correspond to crease structures. For representation purposes sample labels are shortened but should be read SP12-02(letter). (C) Picture taken from the sampling site (see position of SP12-02A) looking East toward the apex of the lava; a large crease structure can be seen in the background (see close up in D). Note that the sampled surface is perfectly smooth though fractured in m-scale compartments and displays stream-parallel striations that are characteristics of pristine crease-structures surfaces. (D) Close-up of the surface sampled for SP12-02A showing ~5-cm wide and few-mm high striations marked on top of the ca. 2-cm-thick rind. (E) Left half-part of the large crease structure situated at the apex of the sampled lava flow showing the general smooth and convex morphology of the rock wall as well as the subhorizontal striations and subvertical wall offsets characteristics of these features. Note the person (for scale) is located in the central valley of the crease structure. The materials forming this structure were exhumed from the central valley up. The second half of this structure can be seen on the right of the person.

Figure 3. $^{40}\text{Ar}/^{39}\text{Ar}$ ages obtained plotted vs. the cumulated amount of ^{39}Ar released for SP12-02A. The calculated plateau ages ($k \pm 1\sigma$) is presented with the MSWD value ($n=8$).

Figure 4. Camel diagram showing the distribution of cosmogenic ^3He concentrations obtained for the San Pedro volcano samples. ^3He concentrations are corrected for magmatic and nucleogenic contributions, scaled for geomorphic shielding and sample thickness and normalized to 21.934°S-68.510°W and 3390 m. Note that normalizing each sample's concentration to the surface, an open-sky topography and to a common coordinate allow to better identifying any potential outliers due to erosion or inheritance. Each individual cosmogenic ^3He concentration is represented as a Gaussian distribution, considering the concentration value and its 1σ -error. Green and dashed-brown curves represent clinopyroxene ($n=5$) and orthopyroxene ($n=3$) aliquots, respectively. Black curve represents the sum of individual curves. The weighted average concentration is calculated after averaging clino- and orthopyroxene values and is reported with its associated MSWD value.

Figure 5. (A) 150-ka-long variations of TCN scaling-factors and magnetic-field cutoff rigidity calculated for 21.934°S-68.510°W-3390 m a.s.l. using calculation procedures and scaling frameworks reported by Martin et al. (2015). All scaling factors are calculated based on the atmospheric pressure derived from the ERA-40 reanalysis dataset (Uppala et al., 2005). Numbers in brackets are the same as in Table 6 and represent predictions from different combinations of scaling schemes and geomagnetic field reconstructions. (B) Time-integrated scaling factors using the “Lm” scaling scheme and the ERA-40 atmosphere model with geomagnetic field reconstructions based on Muscheler-SINT2000 (black curve) and as implemented in the “LSD” model (green curve). The red curves represent the production ratio between “Lm-LSD” and “Lm-Muscheler”.

Figure 6. Distribution of the reevaluated SLHL P3s inferred from the compilation of previous geological calibration sites as well as the San Pedro lava flow. Values represented are inferred from the combination of the “Lm” scaling scheme with the ERA-40 atmosphere model and the geomagnetic field reconstruction relying on the VDM values based on Muscheler et al. (2005) for the period 0-62 ka and SINT2000 (Valet et al., 2005) beyond. (A) Camel diagram showing the distribution of reevaluated SLHL P3s. Inset illustrates the impact of the radiogenic correction applied on cosmogenic ^3He concentrations following Blard and Farley (2008) (see text). (B), (C) and (D) show the distribution of SLHL P3s as a function of latitude, elevation and calibration age, respectively. Note the absence of statistically significant relationships between the production rate and these parameters. Red data represents the San Pedro volcano calibration site.

Figure 1

Figure 2

Weighted mean:
 $99.3 \pm 1.2 \text{ Mat.g}^{-1}$
MSWD = 1.2 (n = 5)

Probability

80 85 90 95 100 105 110 115

$^3\text{He}_c$ concentration (Mat.g $^{-1}$)

Figure 5

Arithmetic mean: $120 \pm 16 \text{ at.g}^{-1}.\text{yr}^{-1}$
 MSWD : 10.4 ($n = 21$)
 Median: $119 \text{ at.g}^{-1}.\text{yr}^{-1}$
 25th perc.: 114; 75th perc.: 130