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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 at.g $^{-1}$.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 at.g $^{-1}$.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

29 **1. Introduction**

30

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

47 The rate at which cosmogenic ^3He is produced at the Earth's surface has been
48 estimated between 100 and 140 $\text{at.g}^{-1}\text{.yr}^{-1}$ (see Dunai, 2010 and reference
49 therein) from several geological calibration datasets with independent age
50 assignments (e.g. Ackert et al., 2003; Amidon and Farley, 2011; Blard et al., 2006;
51 2013; Cerling and Craig, 1994; Dunai and Wijbrans, 2000; Fenton et al., 2013;
52 2014, Foeken et al., 2012; Goehring et al., 2010; Licciardi et al., 1999; 2006). At a
53 global scale, cosmogenic ^3He production rates at sea level and high latitude

54 (SLHL) were estimated using the time-dependent adaptation of the Lal
55 (1991)/Stone (2000) scaling scheme (“Lm”; Balco et al., 2008). The available
56 calibration datasets yielded values of 119 ± 9.9 $\text{at.g}^{-1}\text{.yr}^{-1}$ for pyroxene (Goehring
57 et al., 2010) and 116 ± 13 (Goehring et al., 2010), 122 ± 15 (Blard et al., 2013) and
58 ca. 117 $\text{at.g}^{-1}\text{.yr}^{-1}$ (Borchers et al., 2016) when ^3He concentrations of both
59 pyroxene and olivine minerals are considered. Although the global-scale SLHL
60 cosmogenic ^3He production rates are consistent within associated uncertainties,
61 Phillips et al. (2016) recently emphasized that the initial datasets used in these
62 compilations are clustered both in space (medium to high latitudes) and time (ca.
63 5-20 ka). According to these authors, this circumstance prevents reliable
64 integrations of past atmospheric, geomagnetic and solar modulation conditions
65 that might have affected the production of cosmogenic nuclides at the Earth’s
66 surface. In particular, cosmogenic ^3He calibration data are lacking at latitudes <
67 25° (Blard et al., 2013), where large temporal variations in the paleo magnetic-
68 field intensity might have significantly affected the production of cosmogenic
69 nuclides (e.g. Dunai and Lifton, 2014). Accordingly, improving the accuracy of
70 time dependent corrections requires more low latitude calibration sites with
71 exposure durations significantly longer than 20 ka.

72 This article presents a new calibration of the cosmogenic ^3He production rate
73 from a site located in the hyperarid region of Northern Chile (3400 m a.s.l., ca.
74 22°S), over a long exposure time (ca. 107 ka). The aim is to reduce the lack of
75 long exposure calibration sites located at high elevation/low latitude. Five
76 samples were collected for helium analyses on top of an andesitic lava flow. This
77 surface is characterized by pristine crease-structure features, which are common
78 on silicic volcanic flows and previously identified as reliable targets for surface

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

87

88 **2. Geological setting and sampling**

89

90 *2.1. Volcanic architecture*

91 The San Pedro volcano (6159 m a.s.l., Figure 1) is situated in the El Loa province
92 of Northern Chile ca. 80 km north east of Calama and 100 km north of San Pedro
93 de Atacama. While the San Pedro volcano is located on the western slope of the
94 High Andean Cordillera and therefore does not belong to the Atacama Desert
95 *stricto sensu*, it is characterized by a hyperarid climate, with annual precipitation
96 < 100 mm/y (Houston and Hartley, 2003). The San Pedro volcano is paired with
97 the San Pablo volcano (6118 m a.s.l.) located 5 km further to the east. It
98 represents one of the highest and best-preserved volcanic edifices of the Central
99 Volcanic Zone (de Silva and Francis, 1991), one of the four active volcanic zones
100 in the Andes (see Hora et al., 2007 and reference therein). Only few studies
101 (Francis et al., 1974; Thorpe et al., 1976 and O'Callaghan and Francis 1986)
102 described and identified the nature and composition of erupted materials and
103 the history of eruption of these volcanoes. The San Pedro volcano represents a

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

127

128 *2.2. The SW Dome of the San Pedro volcano*

129 The lava flow emitted by the SW Dome is the largest of the San Pedro volcano. It
130 extends towards the south west for ca. 13 km from ca. 4800 m to 3370 m a.s.l
131 (Figure 1) and covers an area of ca. 20 km² with a volume estimated at 2.8 km³
132 (O'Callaghan and Francis, 1986). The lava flow is considered the product of a
133 200-m-thick early pyroxene dacite lava flow that was locally overlain by
134 subsequent hornblende dacite flows (Figure 1). Although most of the surface of
135 the lava flow displays a rough blocky structure typical of low-viscosity Andean
136 andesite/dacite flows, its distal part presents smooth stream-oriented and
137 elongated features (Figure 2). These features correspond to crease structures
138 (Anderson and Fink, 1992) as inferred from the curved and striated walls
139 situated on both sides of a main central fracture (Figure 2). Such crease
140 structures have been identified as typical constructional volcanic features
141 materializing excellent landforms for surface exposure dating with in-situ
142 produced cosmogenic nuclides (Anderson et al., 1994).

143

144 *2.3. Sampling site*

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

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

158

159 **3. Experimental procedures**

160

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

165

166 *3.1 Sample preparation*

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

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

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

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

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

209

210 *3.2. $^{40}\text{Ar}/^{39}\text{Ar}$ analytical method*

211 About 150 mg of groundmass from SP12-02A was irradiated for 1 hour (IRR-95)
212 in the OSIRIS nuclear reactor (β 1 tube, CEA Saclay, France). After irradiation, ca.
213 40 mg of the sample was transferred into a stainless steel sample holder and
214 then loaded into a differential vacuum Cleartran® window. Argon isotopes were
215 then extracted from the groundmass by step heating using a 25 W CO₂ laser
216 (Synrad®). Ar isotopes were analyzed using a VG5400 mass spectrometer
217 equipped with a single ion counter following the protocol reported in Nomade et
218 al. (2010). Each Ar isotope measurement consisted of 20 cycles of peak jumping
219 over the argon isotopes. Neutron fluence (J) was monitored by co-irradiation of
220 Alder Creek Sanidine (ACs-2, Nomade et al., 2005) placed in the same pit as the
221 sample. J values were determined from analyses of three ACs-2 single grains.
222 Corresponding J value was calculated using an age of 1.194 Ma (Nomade et al.,
223 2005) and the total decay constant reported by Steiger and Jäger (1977). An
224 $^{40}\text{Ar}/^{36}\text{Ar}$ atmospheric ratio of 298.56 was then applied (Lee et al., 2006),
225 consistent with the value reported by Valkiers et al., (2010). Recent revisions of
226 decay and monitor constants suggest values about 0.64 % (Kuiper et al., 2008,
227 1.201 Ma for ACs-2) and 1.0 % (Renne et al., 2011; 1.2059 Ma for ACs-2) older
228 than used here. However, the implied difference in calibrated age is negligible for

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

239

240 *3.3. Helium analyses*

241 Helium analyses were carried out at the CRPG noble-gas lab in Nancy, France.
242 ^3He and ^4He isotope concentrations were measured using the Split Flight Tube
243 mass spectrometer designed by GV. The magmatic contribution to the measured
244 ^3He and ^4He concentrations was estimated by a 6 min in-vacuo crushing (Kurz,
245 1986; Scarsi, 2000) of 97 mg of an aliquot of orthopyroxene of sample SP12-02D.
246 These values were then used for all other samples, assuming that the magmatic
247 contribution is the same in all pyroxenes collected on the San Pedro lava flow.
248 Following Williams et al. (2005) and Blard et al. (2013), we also considered that
249 a potential magmatic contribution is negligible in these micro-phenocrysts.

250 Between ca. 12 and 28 mg of clino- and orthopyroxene were packed in tin foils
251 and fused in-vacuo for 15 min at 1400°C, within an in-house-built single vacuum
252 furnace (Zimmermann et al., 2012). The extracted gas was purified using a
253 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

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

$$281 \quad {}^3\text{He}_c = {}^3\text{He}_t - {}^3\text{He}_n - {}^3\text{He}_m \quad (1)$$

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

300

301 **4. Results**

302

303 *4.1. $^{40}\text{Ar}/^{39}\text{Ar}$ dating of the eruption of the San Pedro Lava flow*

304 The eruption age of the San Pedro volcano lava flow has been estimated from the
305 $^{40}\text{Ar}/^{39}\text{Ar}$ analyses. Results are presented in table 4 and figure 3. The step-
306 heating degassing spectrum yielded a statistically robust plateau age of 107 ± 12 ka
307 (1σ , Mean Square Weighted Deviates, MSWD=0.07) with 100% of the released
308 ^{39}Ar and no rejected heating steps. The age obtained for the total fusion step is
309 consistent with the plateau age at 109 ± 13 ka (Table 4). Alternatively, Ar data
310 were also cast as inverse isochron age. However, this age is poorly constrained at
311 100 ± 52 ka, mainly because of the low radiogenic content in the sample resulting
312 in a very limited spread on the isochron. We therefore discarded this age when
313 calculating the production rates of cosmogenic ^3He . Instead, we applied the
314 $^{40}\text{Ar}/^{39}\text{Ar}$ plateau age of 107 ± 12 ka for the calibration of the cosmogenic ^3He
315 production rate, as this age yields a good reproducibility at all heating steps. This
316 age is also consistent with the exposure age ca. 103 ka reported by Wörner et al.
317 (2002) for the eruption of La Poruña.

318

319 *4.2. Helium isotopic results and corrections applied to ^3He .*

320 Helium isotope analyses are reported in Table 5 and cosmogenic ^3He
321 concentrations ($^3\text{He}_c$) calculated for the San Pedro volcano samples are plotted in
322 Figure 4. Note that helium concentrations are reported in “Mat.g⁻¹” (i.e. 10^6
323 atoms per gram of pyroxene or ca. 3.7×10^{-14} cc STP/g) in the following sections.
324 Total concentrations released by melting ranged between 89 ± 3 and 97 ± 3 Mat.g⁻¹
325 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
326 concentrations cluster around ca. 94 ± 2 Mat.g⁻¹, while the ^4He data show a large
327 scatter, which is probably due to the low amount of released gas and the
328 significant contribution of the furnace blank correction (Table 5). In-vacuo

329 crushing of the aliquot of sample SP12-02D yielded concentrations of
330 (0.29 ± 0.09) Mat.g⁻¹ and $(17.0 \pm 0.6) \times 10^3$ Mat.g⁻¹ for ³He and ⁴He, respectively
331 (Table 5). The gas liberated during crushing has thus a ³He/⁴He isotopic ratio of
332 12 ± 4 Ra that is higher than the values usually reported for volcanic rocks in the
333 Central Andes (Hilton et al., 1993; Blard et al., 2013). This result suggests that
334 unquantifiable amounts of cosmogenic ³He might have been released during in-
335 vacuo crushing, with the consequence that the magmatic correction applied to
336 the samples must be considered as a maximum. However, the impact of this
337 magmatic correction is most likely negligible because the ³He released during
338 crushing (³He_m) only represents ca. 0.3% of the total ³He released during melting
339 (³He_t, Table 5). Note that the magmatic ³He correction applied to all the samples
340 was realized using the ³He concentration measured from the prolonged in-vacuo
341 crushing experiment of the sample SP12-02D. This correction systematically
342 represented less than 0.4% of the total ³He extracted by fusion in all samples and
343 thus does not induce a significant source of uncertainty.

344 Regarding ⁴He, the concentration we measured during the crushing experiment
345 $(17 \pm 0.6) \times 10^3$ Mat.g⁻¹ represents ca. 20% of ⁴He_t determined in the
346 orthopyroxene aliquot of SP12-02D $(77 \pm 23) \times 10^3$ Mat.g⁻¹ (Table 5). If we
347 consider that this ⁴He concentration measured during crushing is magmatic and
348 representative for all samples, then 40 to 80% of ⁴He_t is most likely radiogenic in
349 other samples. Using the radiogenic production rates estimated from the
350 minerals composition (P_{4r} : ca. 1.1 ± 0.1 Mat.g⁻¹.yr⁻¹, Table 3), the calculated (U-Th-
351 Sm)/⁴He ages vary from 12 to 54 ka, i.e. significantly younger than the ⁴⁰Ar/³⁹Ar
352 age of 107 ± 12 ka. Assuming no magmatic ⁴He correction, these (U-Th-Sm)/⁴He
353 ages range from 26 to 76 ka. Such a difference with the ⁴⁰Ar/³⁹Ar age may

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

360 The nucleogenic ^3He ($^3\text{He}_n$) production rates range between ca. 0.1 and 0.2 at.g
361 $^{-1}\text{yr}^{-1}$ (Table 5). The Li concentrations used for this calculation are 20 ppm Li for
362 clinopyroxene and 15 ppm Li for orthopyroxene (from LA ICP-MS analyses of
363 sample SP12-02D, Table 3). Although these values have been used for all 5
364 samples, we emphasize that this simplification does not induce significant
365 uncertainties on the nucleogenic ^3He correction as $^3\text{He}_n$ represents indeed less
366 than 0.03% of $^3\text{He}_t$ (Table 5).

367

368 *4.3. Calculation of cosmogenic ^3He concentrations*

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

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

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

389

390 **5. Discussion**

391

392 *5.1. Estimations of cosmogenic ^3He production rates*

393 The production rate of cosmogenic ^3He can be estimated locally, at the
394 geographic position of the calibration site, and/or scaled to a sea level high
395 latitude (SLHL) reference position. Locally, the cosmogenic ^3He production rate
396 is obtained by dividing the average cosmogenic ^3He concentration obtained for
397 the calibration site by the age of the considered landform deduced from an
398 independent dating technique (e.g. ^{14}C , K/Ar, $^{40}\text{Ar}/^{39}\text{Ar}$). Note that individual
399 concentrations are first normalized to the surface, an open-sky topography and
400 the mean geographic coordinates (latitude, longitude and elevation) in order to
401 identify potential outliers due to erosion or inheritance (see e.g. Martin et al.,
402 2015, and reference therein). Scaling the cosmogenic ^3He production rate to
403 SLHL (referred to as SLHL P3 in following sections) is, however, not

404 straightforward. The scaling procedure indeed requires the combination of a
405 geographic spatialization model with an atmosphere model and the
406 reconstruction of the past geomagnetic field conditions (see Balco et al., 2008;
407 Lifton et al., 2014). Here we followed a calculation procedure based on a least-
408 square minimization technique (see Martin et al., 2015, for details) and the
409 scaling frameworks presented bellow. SLHL P3 estimates were inferred from the
410 San Pedro samples using the time-independent Lal/Stone scaling model (Lal,
411 1991; Stone, 2000), noted “St”, as well as two time-dependent schemes: “Lm”, the
412 adaptation of the Lal/Stone model (Balco et al., 2008) and “LSD”, recently
413 developed by Lifton et al. (2014). SLHL P3 estimates were thus obtained by
414 dividing the local production rate by a scaling factor representative for a given
415 combination of scaling, atmosphere and geomagnetic reconstruction models.
416 Regarding scaling schemes, we favored the “Lm” and “LSD” time-dependent
417 models. Indeed, recent studies (Lifton et al., 2014; Borchers et al., 2016; Phillips
418 et al., 2016) have demonstrated that these scaling schemes yield more consistent
419 and reliable spatial and temporal estimates of cosmogenic nuclide production
420 rates than former neutron-monitor based models (Desilets and Zreda, 2003;
421 Desilets et al., 2006; Dunai, 2001; Lifton et al., 2005). These “Lm” and “LSD”
422 models thus represent the most widely used and recently available scaling
423 schemes, respectively (see e.g. Lifton et al., 2014 and Martin et al., 2015). We
424 then calculated SLHL P3s for “St”, “Lm” and “LSD” using the Standard
425 atmosphere model (National Oceanic and Atmospheric Administration, 1976)
426 and the ERA-40 reanalysis dataset (Uppala et al., 2005), which allows accounting
427 for the regional variations in height-pressure relationship. Finally, we inferred
428 SLHL P3s for the time-dependent “Lm” and “LSD” scaling schemes with three

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

437

438 *5.2. Calibration of $^3\text{He}_c$ production rates at 21.934°S-68.510°W-3390 m a.s.l.*

439 The local cosmogenic ^3He production rate, estimated for the San Pedro
440 calibration site at a geographic position of 21.934°S-68.510°W and 3390 m a.s.l.,
441 is obtained by dividing the weighted average $^3\text{He}_{c(s)}$ with the eruption age of the
442 San Pedro lava flow obtained from the $^{40}\text{Ar}/^{39}\text{Ar}$ dating. This approach yields a
443 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
444 propagating both the cosmogenic ^3He and $^{40}\text{Ar}/^{39}\text{Ar}$ uncertainties respectively, at
445 the 1-sigma level (Table 5). This rate combines all production pathways of
446 cosmogenic ^3He including spallation, thermal neutron capture and muogenic
447 production. We note however, that the contribution of cosmogenic thermal
448 neutrons to the production of cosmogenic ^3He at the San Pedro calibration site is
449 likely lower than 5% and therefore negligible, based on the predictions obtained
450 with the model reported by Amidon and Farley (2011) for pyroxenes with 15-20
451 ppm apparent Li and a He-closure age of ca. 100 ka.

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

454 samples are presented in Table 6. SLHL P3s obtained from the time-independent
455 “St” scheme with the Standard and ERA-40 atmosphere models are 132 ± 14 and
456 143 ± 16 $\text{at.g}^{-1}\text{.yr}^{-1}$, respectively. SLHL P3s estimated with the time-dependent
457 “Lm” and “LSD” schemes, with various combinations of atmosphere and
458 geomagnetic models, vary from 103 ± 11 to 130 ± 14 $\text{at.g}^{-1}\text{.yr}^{-1}$ (Table 6). In
459 general, the inferred values agree well with the SLHL P3 reported from global-
460 scale compilations by Goehring et al. (2010) and more recently by Blard et al.
461 (2013a). Moreover, SLHL P3s that we estimated using the time-dependent
462 adaptation of the Lal/Stone scaling scheme (“Lm”) with the Muscheler-SINT2000
463 geomagnetic reconstruction and considering both the Standard and ERA-40
464 atmosphere models are 120 ± 13 and 130 ± 14 $\text{at.g}^{-1}\text{.yr}^{-1}$, respectively.
465 Interestingly, these SLHL P3 estimates fit perfectly with the values reported by
466 Blard et al. (2013a) from a ca. 15-ka-old calibration site located at 3800 m a.s.l.
467 on the Bolivian Altiplano (Figure 1).

468 Note that all applied time-dependent scaling frameworks predict that at the San
469 Pedro volcano calibration site, the production of cosmogenic nuclides, including
470 cosmogenic ^3He , has been generally higher during the last 100 to 150 ka than at
471 present (Figure 5). Such a situation can be explained by the persistence of a
472 weaker geomagnetic field intensity that has induced lower cutoff rigidities,
473 thereby allowing more cosmic-ray particles to enter the Earth’s magnetic field
474 (see Dunai and Lifton, 2015 and references therein). This effect is particularly
475 amplified in high tropical areas integrating >50-ka exposure histories (e.g. Balco
476 et al., 2008), since it yields relative corrections between 10 and 20% of the
477 modern rate (Figure 5). It thus explains why the San Pedro calibration site yields
478 time-independent “St” scaling factors that are higher than those inferred from

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

486

487 *5.3. Re-evaluation of the global SLHL cosmogenic ³He production rate*

488 Cosmogenic ³He production rates have rarely been calibrated to sea level high
489 latitude positions using a common calculation framework that included similar
490 scaling schemes, atmosphere model and geomagnetic field reconstructions. Here
491 we re-evaluate the global SLHL P3 by compiling and processing available
492 calibration datasets for both olivine and pyroxene within a common calculation
493 framework that combines the scaling schemes, atmosphere and geomagnetic
494 field models presented above (see section 3.4). Twenty different calibration sites
495 and ca. 150 cosmogenic ³He analyses have been compiled from the literature and
496 combined with the San Pedro samples (see Table A2). This new compilation
497 includes the studies of Ackert et al. (2003), Amidon and Farley (2011), Blard et
498 al. (2006, 2013a), Cerling and Craig (1994); Dunai and Wijbrans (2000), Fenton
499 et al. (2013, 2014), Foeken et al. (2012), Goehring et al. (2010) and Licciardi et al.
500 (1999, 2006). We do note that we only considered dataset originating from
501 absolute calibration studies and therefore excluded those from cross calibrations
502 (e.g. Amidon et al, 2009). Some of the studies included in this compilation did not
503 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 $76\pm 4 - 186\pm 12 \text{ at.g}^{-1}\text{.yr}^{-1}$
519 $^1\text{.yr}^{-1}$ (see Table A2). When considering each scaling framework separately, the
520 values yield arithmetic means that range between 115 ± 19 and $122\pm 20 \text{ at.g}^{-1}\text{.yr}^{-1}$,
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

529 $120 \pm 16 \text{ at.g}^{-1}\text{.yr}^{-1}$. Figure 6 represents the distribution of SLHL P3s inferred from
530 this latter scaling framework. It does not show any correlation with elevation nor
531 with latitude. This confirms previous observations reported by Goehring et al.
532 (2010) and Blard et al. (2013a) that there is no anomalous production of
533 cosmogenic ^3He at high elevations, at least in pyroxene and olivine. It is,
534 however, important to emphasize that the estimated Mean Square Weighted
535 Deviation is significantly higher than 1 (MSWD = 10.4; Figure 6), indicating that
536 the observed variability in the data is larger than a statistical dispersion that
537 would result from analytical uncertainties only. In other words, the global SLHL
538 P3 compilation unlikely represents a single population. Note that while Figure 6
539 only represents SLHL P3s estimated from the combination of the “Lm” scaling
540 scheme with the ERA-40 atmosphere model and Muscheler-SINT2000 based
541 geomagnetic field reconstruction, all other scaling frameworks present a similar
542 picture. Part of this over-dispersion might result from calibration sites older than
543 ca. 120 ka that systematically yield SLHL P3s lower than ca. $110 \text{ at.g}^{-1}\text{.yr}^{-1}$ when
544 considering the “Lm” scaling scheme (Figure 6). This inference, however, should
545 be tuned down when considering the “LSD” scaling scheme, as P3s estimates do
546 not show such temporal pattern. In any cases and for all considered scaling
547 frameworks, the variability in SLHL P3s seems independent from the exposure
548 age for calibration sites younger than 110 ka (Figure 6). The observed variability
549 in SLHL P3s might result from unrecognized erosion, variations in atmospheric
550 pressure, inaccurate correction of the time-dependent production rate
551 variability, and/or unaccounted excess in Li concentrations of crustally
552 contaminated magmas (e.g. Amidon et al., 2011; Dunai and Wijbrans, 2000;
553 Dunai et al., 2007; Foeken et al., 2012; Staiger et al., 2007). Excluding calibration

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

567

568 **6. Conclusion**

569

570 We report a new geological calibration site located in northern Chile (21.934°S-
571 68.510°W) at 3390 m a.s.l. that we used to infer the production rate of
572 cosmogenic ^3He in pyroxenes. $^{40}\text{Ar}/^{39}\text{Ar}$ dating gives a consistent plateau age of
573 107 ± 12 ka. Eight mineral separates of clino- and orthopyroxene from five
574 different surface samples yield clustered ^3He concentrations, for both pyroxene
575 phases. Magmatic and nucleogenic ^3He corrections are negligible ($< 1\%$) in this
576 lava flow. The weighted average cosmogenic ^3He concentration is 99.3 ± 1.2 Mat.g⁻¹,
577 which represents the highest cosmogenic ^3He concentrations ever measured so
578 far in natural calibration samples.

579 We then used this data to constrain a local cosmogenic ^3He production rate of
580 928 ± 101 at.g $^{-1}$.yr $^{-1}$ at 21.934°S-68.510°W and 3390 m a.s.l. After scaling to sea
581 level and high latitudes (SLHL), cosmogenic ^3He production rate estimates vary
582 from 103 ± 11 to 130 ± 14 at.g $^{-1}$.yr $^{-1}$, which depends on different combinations of
583 scaling schemes, atmosphere models and geomagnetic field reconstructions.
584 Finally we combined the SLHL P3s inferred from the San Pedro lava flow
585 samples with cosmogenic ^3He production rates (in both olivine and pyroxene)
586 inferred from various calibration sites and re-evaluated the mean global SLHL
587 P3. This compilation, where cosmogenic ^3He production rates were scaled to
588 SLHL with two scaling schemes, two atmosphere models and three different
589 geomagnetic field reconstructions, yield arithmetic means for SLHL P3s that vary
590 between 115 ± 19 and 122 ± 20 at.g $^{-1}$.yr $^{-1}$. These values are identical within
591 uncertainty and consistent with values previously reported from similar
592 compilation studies (e.g. Goehring et al., 2010; Blard et al., 2013a; Borchers et al.,
593 2016). This new compilation thus illustrates that a calculation framework
594 including the (1) time-dependent adaptation of the Lal (1991) / Stone (2000)
595 scheme, (2) ERA-40 atmosphere model (Uppala et al., 2005), and a (3)
596 geomagnetic field reconstruction relying on the Virtual Dipole Moment values
597 based on Muscheler et al. (2004) and SINT-2000 (Valet et al., 2005) currently
598 yields the best constrained mean global-scale cosmogenic ^3He production rate.
599 Our observations also appear perfectly in line with the recent findings reported
600 by Lifton (2016). We thus suggest that the update of the scaling framework
601 presented here, together with the inferred mean global SLHL P3 of 120 ± 16 at.g $^{-1}$.yr $^{-1}$,
602 should be favored when calculating exposure ages from cosmogenic ^3He
603 concentrations.

604

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606

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613

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615

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Table 1. Sample characteristics

Sample name	Latitude (°)	Longitude (°)	Elevation (m)	Atmospheric pressure (hPa) ^a	Altitude factor ^b	Sample thickness (cm)	Density (g.cm ⁻³)	Thickness correction ^c	Geomorphic shielding ^d	Total correction ^e (%)
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 (ppm)	B (ppm)	Cr (ppm)	Co (ppm)	Ni (ppm)	Gd (ppm)	U (ppm)	Th (ppm)	Sm (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 $(3.3\pm 1.1) \times 10^4$ atoms and $(1.8\pm 0.4) \times 10^9$ 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 $(17.0\pm 0.6) \times 10^3$ 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 1σ -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 ($k\alpha \pm 1\sigma$) 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

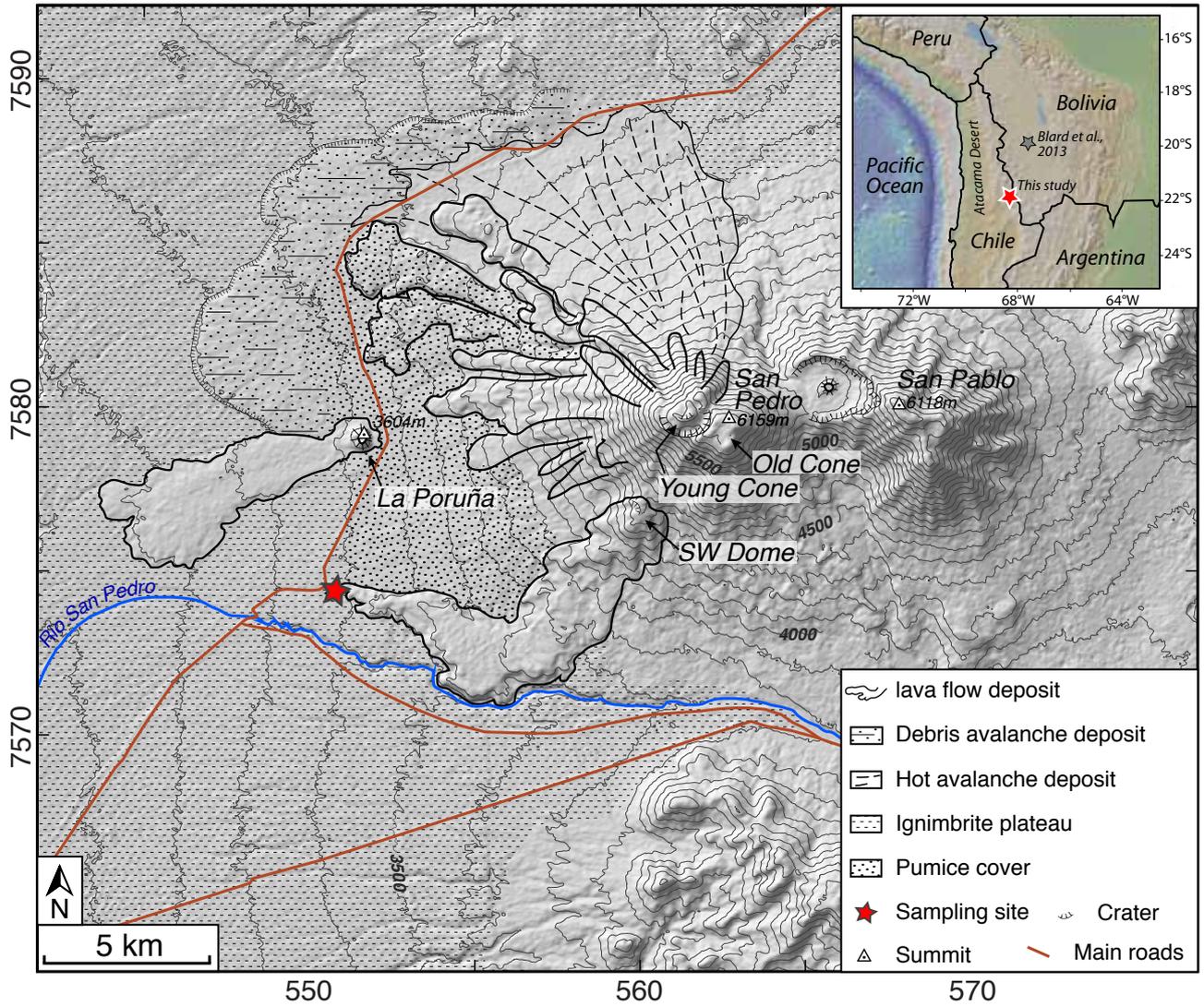
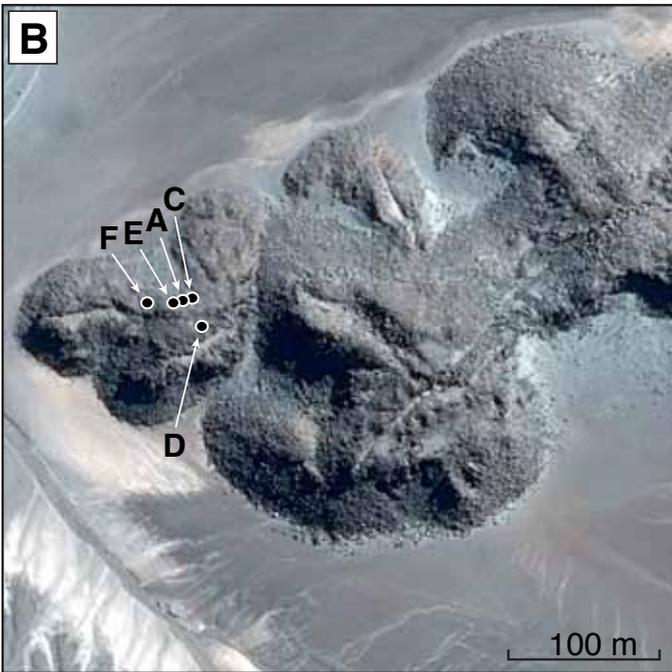
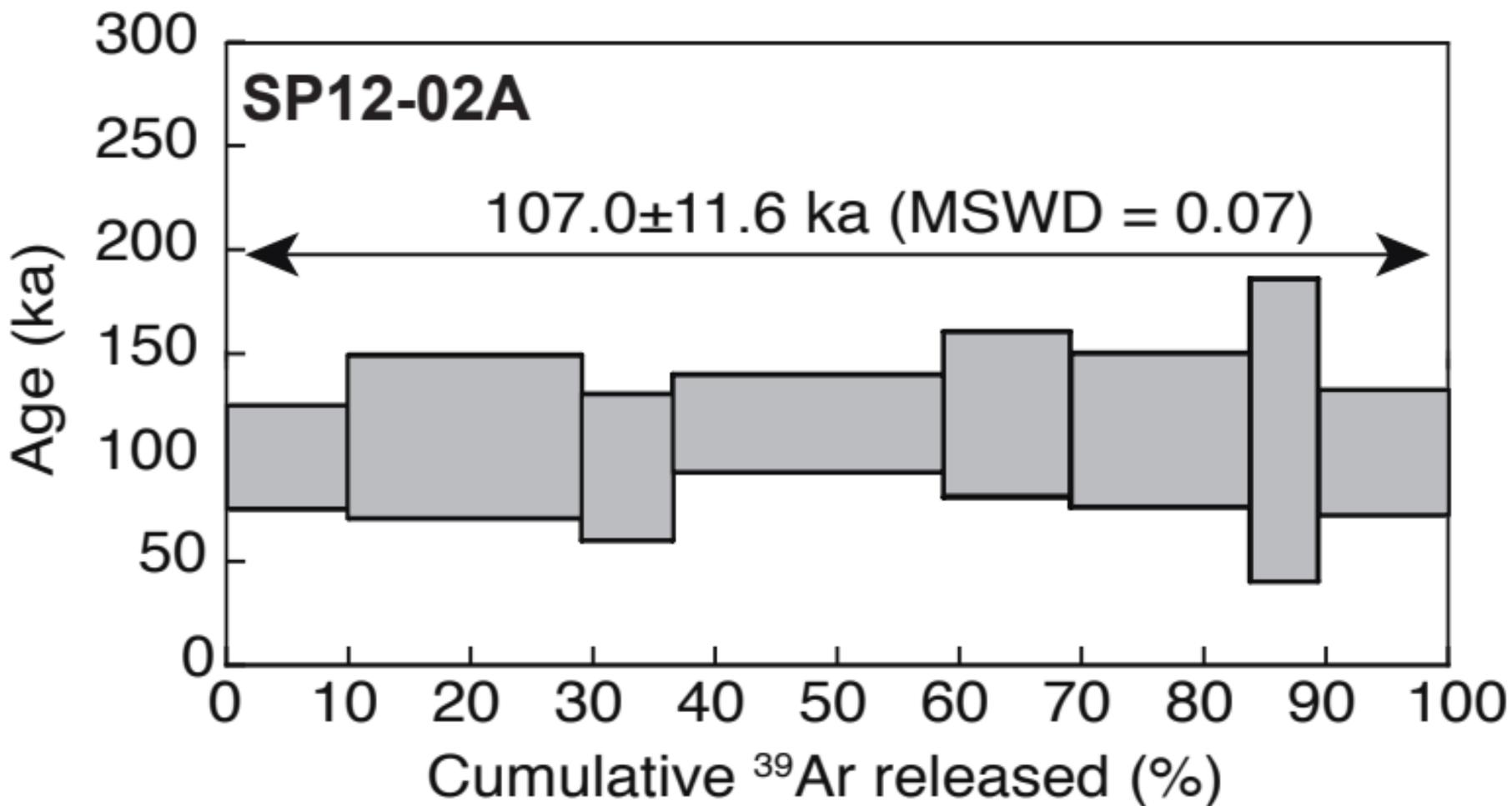


Figure 2





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 $^{-1}$)

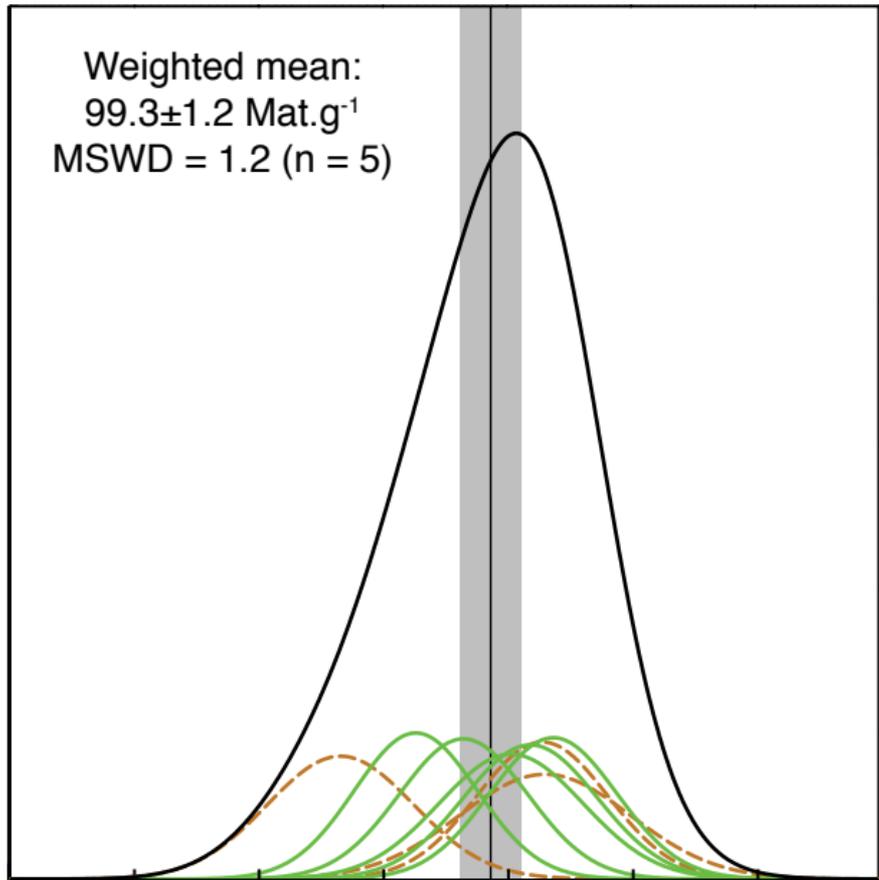
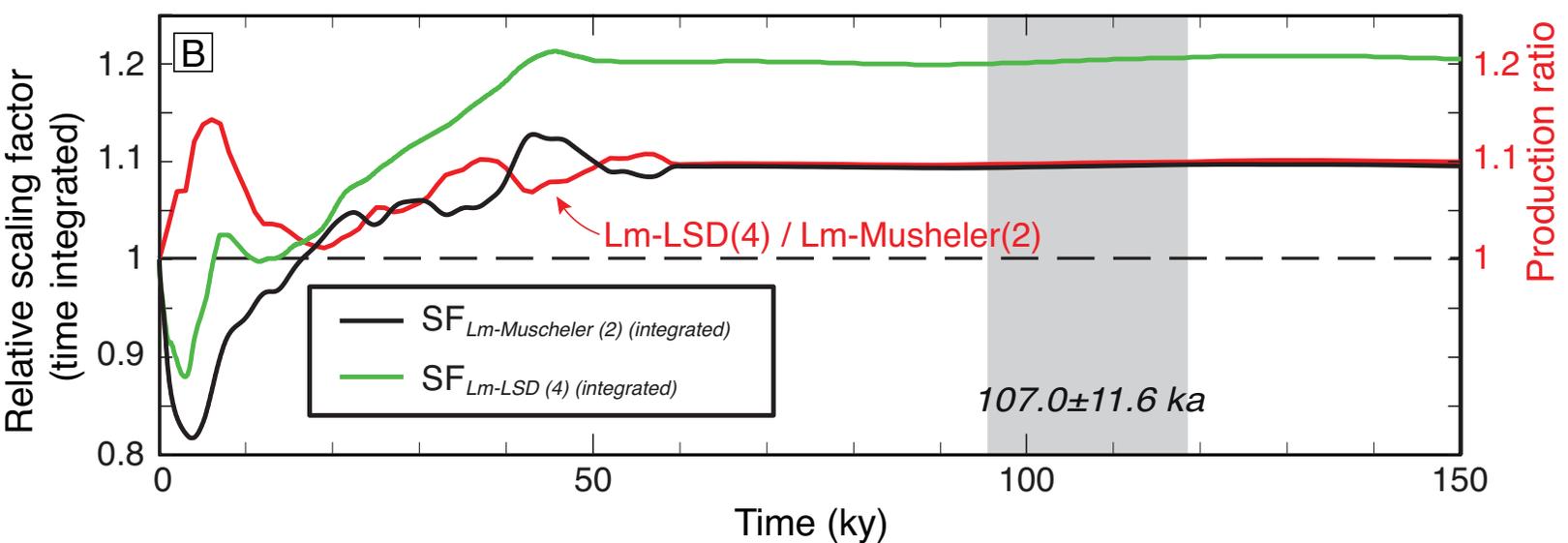
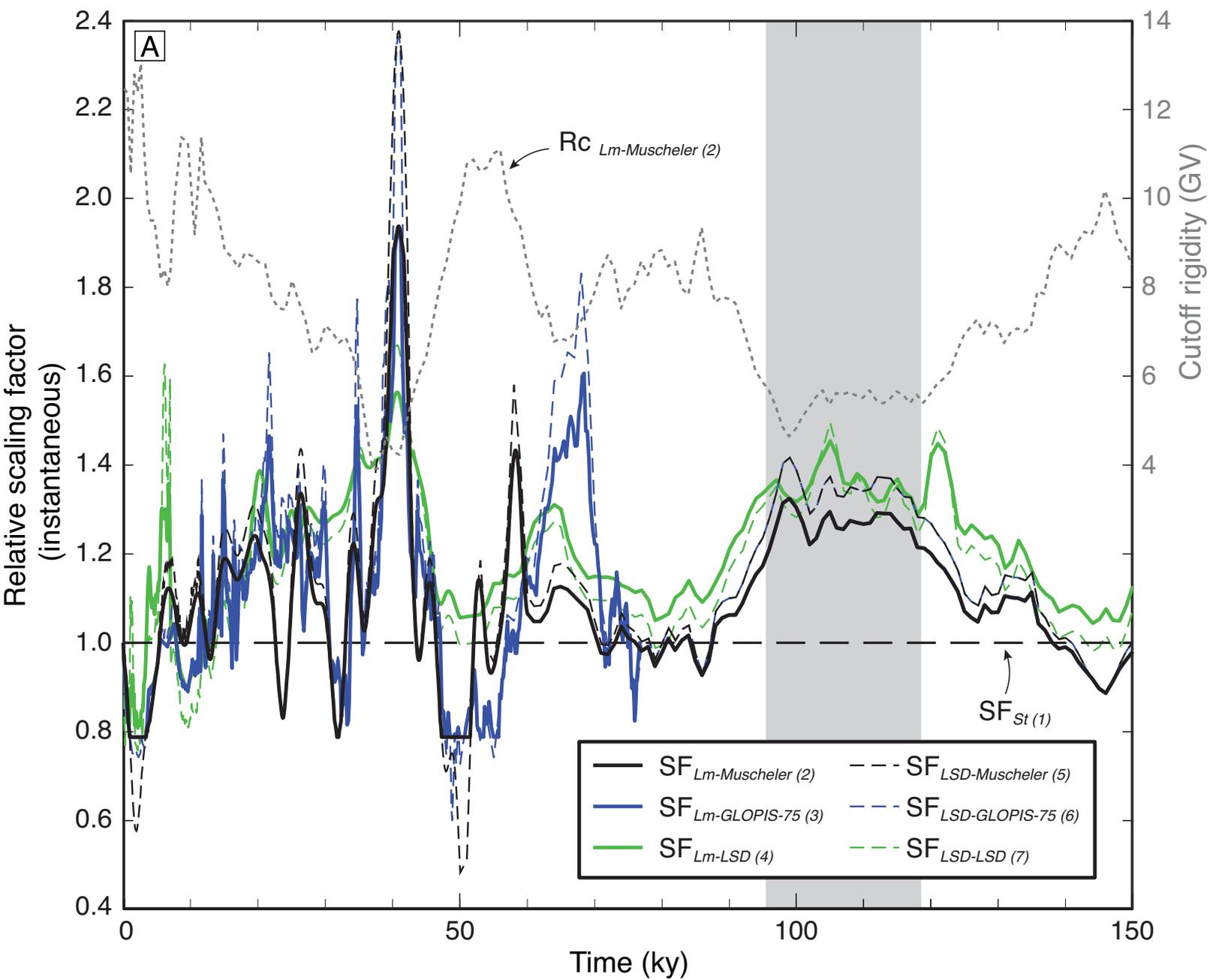
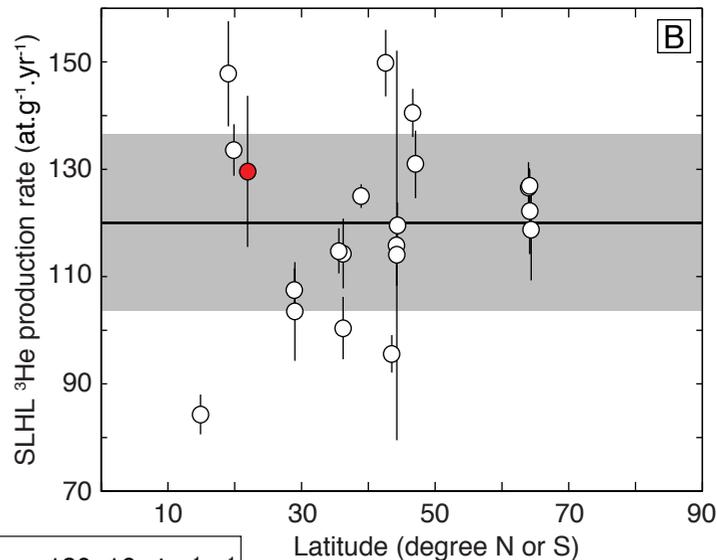
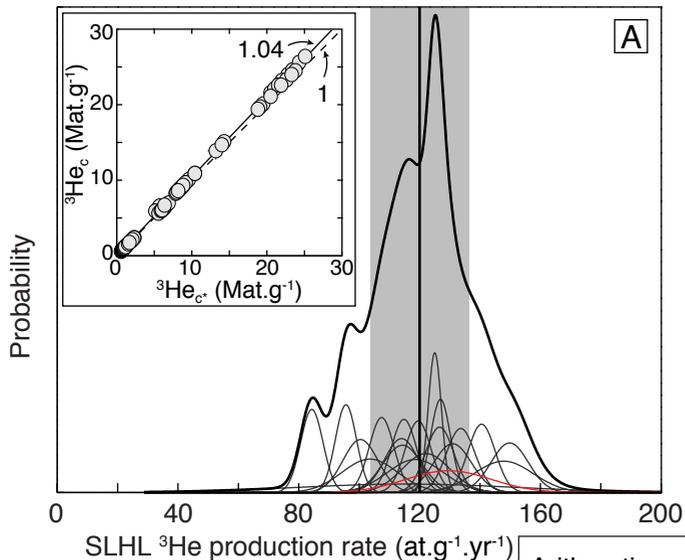


Figure 5





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

