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3	Temperatures since the last glacial maximum in the Maritime Alps
4	from cosmogenic noble gas paleothermometry
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# 24 Abstract

While an extensive network of proxy records has been used to reconstruct climate parameters 25 throughout much of the European Alps during the Late Quaternary, our knowledge of deglacial 26 climate conditions in the Maritime Alps from proxy records is limited. Here, we utilize a new 27 geochemical technique known as cosmogenic noble gas paleothermometry to constrain 28 temperatures in the Maritime Alps since the Last Glacial Maximum (LGM). We measured 29 cosmogenic <sup>3</sup>He abundances in guartz from boulders in a series of nested moraines in the Gesso 30 Valley, Italy. Paired with previous constraints on the exposure duration from cosmogenic <sup>10</sup>Be 31 measurements and with <sup>3</sup>He diffusion experiments on proton-irradiated quartz fragments from 32 the same boulders, we use the cosmogenic <sup>3</sup>He abundances to place constraints on the possible 33 temperatures these boulders experienced during their exposure since the LGM. We calculate 34 effective diffusion temperatures over the last ~22 thousand years that range from 8 to 25 °C. 35 Effective diffusion temperatures (EDTs) are functionally related to but generally several degrees 36 higher than mean ambient temperatures due to the nonlinear dependence of diffusion on 37 temperature. The EDTS we determine from our <sup>3</sup>He observations are broadly consistent with 38 temperatures inferred from other proxies in nearby Alpine regions and with temperature 39 estimates from a general circulation model (GCM). In detail, however, we calculate significantly 40 different EDTs for boulders sampled from the same moraines. These discrepancies could result 41 from (1) the fact that we have not accounted for variations in radiative heating of the boulder 42 surfaces (e.g., due to variations in overhead vegetation cover or snow cover), (2) limitations in 43 our understanding of and ability to model accurately complex helium diffusion kinetics, (3) 44 uncertainties associated with our grain size analyses, or (4) some combination of the three. 45 46 Additional work to understand the effects of radiative heating and to better understand complex

diffusion kinetics is necessary to provide better resolution on temperatures since the LGM in thissetting.

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#### 50 **1. Introduction**

Data from paleoclimate proxies constrain the dynamics of Earth's climate system on timescales 51 inaccessible with the instrumental record. Proxy observations from key intervals of Earth's 52 climatic past, such as the Last Glacial Maximum (LGM) and subsequent deglaciation, allow us 53 to understand how the climate system responds to a change in forcing (e.g., Annan and 54 Hargreaves, 2013; Schmittner et al., 2011; von der Heydt et al., 2016) and evaluate the ability of 55 climate models to simulate the climate system's response (e.g., Annan and Hargreaves, 2015; 56 Harrison et al., 2015; Schmidt et al., 2014) both of which improve our ability to forecast future 57 climate change (e.g., Masson-Delmotte et al., 2013). Terrestrial proxies are particularly 58 important for assessing how changes in large scale ocean-atmosphere dynamics manifest at a 59 regional scale in the continental settings that people inhabit and depend upon. 60

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An extensive network of terrestrial climate reconstructions from fossil pollen and chironomid 62 (midges) records exists across the European Alps since the Last Glacial Maximum (LGM; e.g. 63 Bartlein et al., 2011; Heiri et al., 2014). Despite such an extensive proxy network, our knowledge 64 of deglacial climate conditions in the southern sector of the French and Italian Alps (hereafter 65 66 referred to as the Maritime Alps) from proxy records is limited. While there are a handful of pollen-based reconstructions from the Maritime Alps of climate parameters through the 67 Holocene, such as mean temperature during the coldest month and mean annual precipitation 68 69 (Bartlein et al., 2011; Cheddadi et al., 1996; Cheddadi and Bar-Hen, 2009; Davis et al., 2003;

Wu et al., 2007), no reconstruction from the region extend back to the LGM. Similarly, there is 70 only one reconstruction of mean July temperatures from fossil assemblages of chironomids 71 proximal to the Maritime Alps from Lago Piccolo di Avigliana with temporal coverage limited to 72 14–9.5 thousand years ago (Larocque and Finsinger, 2008). There are two proxy studies from the 73 Maritime Alps that extend into the latest Pleistocene which we are aware of-one reporting 74 pollen fossil assemblages in sixteen high elevation (> 1700 m) lakes (Brisset et al., 2015) and 75 one reporting fossil chironomid assemblages from Lac Long Inférieur in France (Gandouin and 76 Franquet, 2002)—and neither study interprets the proxies in terms of quantitative climate 77 78 parameters.

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Due to the Maritime Alps' (1) southerly latitude, (2) generally lower elevations and (3) proximity 80 to the Mediterranean Sea, the Late Quaternary climatic evolution and glacier dynamics in this 81 region may have differed substantially from those in Alpine regions further north and further 82 inland (e.g., Kuhlemann et al., 2008), despite the apparent synchronicity of glacier advances 83 during the LGM and later stadials recorded by moraines across the Alps (Federici et al., 2017; 84 e.g. Ivy-Ochs et al., 2007; Kuhlemann et al., 2008). Reconstructing temperatures in the Maritime 85 Alps since the LGM would therefore fill a gap in an otherwise extensive network of proxy-based 86 reconstructions of post-LGM climate across the Alps, and address the specific question of how 87 climate evolution in the Maritime Alps during deglaciation may have differed from other Alpine 88 89 sectors. For example, in combination with the positions and chronology of moraines, a wellresolved local temperature record could be used to inform a simple glacier mass balance model 90 for valleys in the Maritime Alps during the deglacial period. With independent constraints on 91 92 deglacial temperatures, such a mass balance model could be used to crudely invert for changes in

precipitation during deglaciation (e.g. Kessler et al., 2006). This paired temperature-precipitation
reconstruction could then be compared to proxy records further north and inland in the Alps to
assess how climate and glacier dynamics varied across the region during deglaciation.

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In addition to assessing climatic variability during deglaciation across the European Alps, 97 temperature records from an independent proxy could provide needed tests of different 98 methodologies for reconstructing climate parameters over this time interval. Reconstructions of 99 climate parameters from existing proxies in the European Alps commonly exhibit significant 100 101 disagreement with climate model simulations prior to the Holocene. Pollen-based reconstructions suggest much colder temperatures, particularly in the winter, than the models predict (e.g. Jost et 102 al., 2005), while models predict higher summertime temperatures than chironomid-based 103 reconstructions during interstadials (Heiri et al., 2014). It is unclear whether these disagreements 104 are attributable to limitations in the global climate models used to simulate paleoclimate, a 105 breakdown in the assumptions of biologically-based reconstructions, or some combination of 106 both. Having independent pre-Holocene temperature records in the European Alps that are 107 mechanistically governed by different processes would therefore help in assessing the cause of 108 the current proxy-model discrepancies. 109

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111 Cosmogenic noble gas paleothermometry is a new geochemical technique for reconstructing past 112 Earth surface temperatures (Tremblay et al., 2014a). Cosmogenic nuclides are produced by 113 nuclear interactions between target atoms and high-energy cosmic-ray particles, both in Earth's 114 atmosphere and in the solid Earth. Because the flux of secondary cosmic-ray particles decreases 115 exponentially with depth in rock, production of cosmogenic nuclides is restricted to the

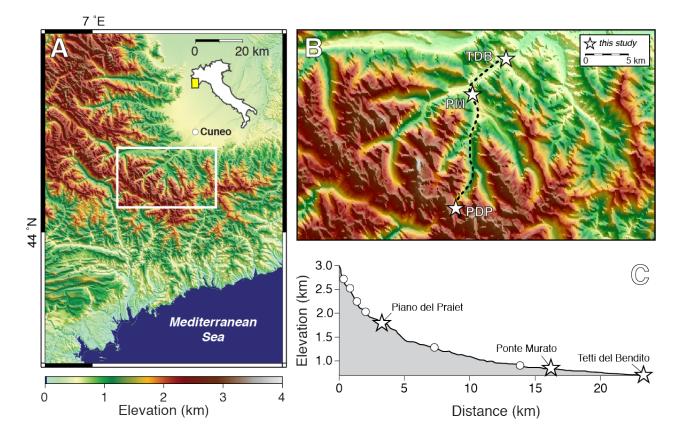
uppermost few meters of the Earth's crust. The near surface production of rare nuclides such as 116 <sup>10</sup>Be and <sup>26</sup>Al by cosmic-ray particle interactions in common rock-forming minerals like guartz 117 has been extensively developed and utilized by the geomorphology community to date the 118 119 surface exposure of rocks and quantify erosion rates over the last 25 years (e.g., Granger et al., 2013). Rare noble gas nuclides such as <sup>3</sup>He and <sup>21</sup>Ne are also produced in minerals by cosmic-120 ray particle interactions. In common minerals like quartz and feldspars, these noble gases 121 sometimes exhibit diffusive loss at Earth surface temperatures (Gourbet et al., 2012; Shuster and 122 Farley, 2005; Tremblay et al., 2017, 2014b) and thus have not been widely utilized in 123 cosmogenic nuclide studies. However, cosmogenic noble gas-mineral pairs exhibiting open-124 system behavior (simultaneous production and diffusion) can be utilized to reconstruct the 125 temperatures that rocks experience while exposed to cosmic-ray particles at Earth's surface when 126 paired with observations of a quantitatively retained cosmogenic nuclide. In high latitude and/or 127 high altitude environments, theoretical calculations indicate that measurements of <sup>3</sup>He in guartz 128 can record temperature variations from the last glacial period to the present (Tremblay et al., 129 2014a). 130

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Here, we constrain temperatures since the last glacial maximum (LGM) using observations of cosmogenic <sup>3</sup>He abundances in quartz from moraine boulders in the Gesso Valley. The Gesso Valley is located in the southernmost Maritime Alps of Italy approximately 40 km from the Gulf of Genoa in the Mediterranean Sea (Fig. 1) and was glaciated during the last glacial period. Glacial moraines and other geomorphic features have been mapped throughout the basin (Federici et al., 2003), and cosmogenic <sup>10</sup>Be exposure ages have been determined for boulders from three moraines that span from the LGM to the Younger Dryas cold period (YD) (Federici et

al., 2017, 2012, 2008). A number of younger, Holocene-age moraines are preserved at higher 139 elevations in the Gesso Valley (Fig. 1), and small glaciers restricted to the highest elevation 140 cirques persist today (Federici et al., 2017). We measured cosmogenic <sup>3</sup>He abundances in quartz 141 from a subset of the boulders with <sup>10</sup>Be exposure ages from the three previously-studied 142 moraines. We also conducted stepwise degassing experiments to quantify the kinetics of <sup>3</sup>He 143 diffusion in guartz from these boulders. Because cosmogenic <sup>3</sup>He is simultaneously produced 144 and diffusively lost during exposure in the moraines, the 'apparent' exposure ages we calculate 145 from the measured <sup>3</sup>He abundances are younger than the 'true' exposure age determined from the 146 <sup>10</sup>Be measurements. The difference between the 'apparent' and 'true' exposure ages can be used 147 to calculate the integrated effective diffusion temperature (EDT) during exposure. This 148 integrated EDT is the temperature corresponding to the mean diffusivity over a variable 149 temperature history, and is therefore a direct function of the ambient temperature history 150 (Tremblay et al., 2014a). Paired with sample-specific <sup>3</sup>He diffusion kinetics, we use the 151 difference between 'apparent' and 'true' exposure ages to model the permissible integrated 152 EDTs, and by relation ambient temperature histories, of each boulder. The presence of numerous 153 moraines with different exposure ages within the same valley is advantageous because it allows 154 us to reconstruct temperatures over different time intervals for an area with a shared climate 155 history. 156

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**Figure 1:** A: Topographic map of the Maritime Alps. Map location is shown in the inset of Italy. White box corresponds to the enlarged view in (B). B: Topography of the Gesso Valley. White stars mark the locations of moraines sampled for this study: Piano del Praiet (PDP), Ponte Murato (PM), and Tetti del Bandito (TDB). Dashed line corresponds to the longitudinal profile shown in (C). C: Longitudinal profile of the Gesso Valley. White stars mark the locations of moraines studied here, as in (B); white circles denote additional moraines in the Gesso Valley not studied here. A detailed map of the geomorphic features in the Gesso Valley is available in Federici et al. (2003).

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## 160 **1.1 Existing proxy constraints on the Late Quaternary climate of the Maritime Alps**

161 Over the historical period, the Maritime Alps have been characterized by warmer mean annual 162 temperatures (MAT), smaller annual temperature amplitudes, lower mean annual precipitation 163 (MAP), and snow cover that is thinner and lasts for smaller fraction of the year than in other 164 Alpine sectors (Durand et al., 2009a, 2009b). The Maritime Alps most likely remained the 165 warmest Alpine sector during the Late Quaternary, given that the Maritime Alps are  $\sim$ 6° south of 166 the northernmost eastern Alps, and thus would have experienced greater insolation and generally higher temperatures during this time period. Warmer overall conditions is consistent with species 167 distribution and phylogeographical models, which suggest that the Maritime Alps remained 168 warmer than other parts of the Alps and functioned as a refugium for plant species during the last 169 glacial period (Casazza et al., 2016). However, temperature and precipitation anomalies in the 170 Maritime Alps during specific climatic intervals of the Late Quaternary may have been distinct 171 from other Alpine sectors. For example, during the Mid-Holocene Warm period 6,000 years ago, 172 pollen-based reconstructions indicate that Maritime Alps winters may have been as much as 2–3 173 °C colder than today, while throughout the rest of the Alps winter temperatures were comparable 174 to today (Bartlein et al., 2011; Cheddadi et al., 1996; Wu et al., 2007). For the same time period, 175 pollen-based reconstructions indicate that the Maritime Alps were wetter ( $\Delta P - E > 0$ ), while the 176 rest of the Alps were drier ( $\Delta P - E < 0$ ) (Bartlein et al., 2011; Cheddadi et al., 1996). 177

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As previously mentioned, no proxy-based reconstructions from the Maritime Alps extend back to 179 the LGM. The nearest pollen-based reconstructions indicate 15–20 °C colder winter temperatures 180 and ~500 mm/yr lower MAP during the LGM than today (Bartlein et al., 2011; Jost et al., 2005). 181 182 For the Mediterranean region as a whole, LGM winter temperatures range anywhere from 0 to 30 °C colder than today, while MAP varies between 0 and 800 mm/yr less than today (Wu et al., 183 2007). In contrast to pollen-based reconstructions, Kuhlemann et al. (2008) suggested that 184 185 incursions of cold polar air between the Pyrenees and the Alps were more frequent during the LGM in order to explain spatial patterns of Mediterranean sea surface temperatures (SSTs) and 186 equilibrium line altitude (ELA) depressions. Such cold air incursions would have resulted in 187 188 greater moist air convection and cyclogenesis as these air masses encountered the warm SSTs in

the Gulf of Genoa and therefore greater precipitation in the Maritime Alps and other Mediterranean-proximal glaciated areas (e.g., Corsica). Although this interpretation of Kuhlemann et al. (2008) is complicated by the fact that the ELA dataset they used is diachronous (Hughes and Woodward, 2017), it nonetheless implies wetter conditions in the Maritime Alps, not dryer as suggested by pollen-based reconstructions, during the LGM and potentially during subsequent stadials marked by glacier re-advances, assuming the same atmospheric phenomena took place.

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#### 197 **2. Methods**

Of the Gesso Valley boulders for which <sup>10</sup>Be measurements exist, we obtained archived material 198 (either whole rock or crushed, sieved fractions) from five samples for cosmogenic <sup>3</sup>He 199 measurements: one from the Piano del Praiet (PDP10) moraine, two from the Ponte Murato 200 moraine (PM1, and PM4), and two from the Tetti del Bandito moraine (TDB1 and TDB3; Fig. 1 201 and Table 1). The <sup>10</sup>Be concentration in quartz for PDP10 (223.8  $\pm$  9.6  $\times$  10<sup>3</sup> atoms/g) was 202 measured at the Scottish Universities Environmental Research Centre Cosmogenic Isotope 203 Analysis Facility in 2013; all other <sup>10</sup>Be concentrations are previously published (Federici et al., 204 2012, 2008). The <sup>10</sup>Be exposure ages of these boulders and other boulders from the same 205 moraines have been used to associate the Piano del Praiet, Ponte Murato, and Tetti del Bandito 206 moraines with the Younger Dryas, Buhl, and LGM stadials respectively (Federici et al., 2012, 207 2008). We obtained a seventh sample, PDP-11, for which there are no existing <sup>10</sup>Be 208 measurements. However, probative measurements of neon isotope abundances in PDP-10 and 209 PDP-11 indicated that PDP-11 has a substantial inherited cosmogenic nuclide inventory (i.e., 210 prolonged exposure to cosmic rays before being deposited in the Piano del Praiet moraine) and 211

will therefore not be discussed further. We separated quartz from other phases using standard 212 crushing, sieving, and magnetic methods, followed by a 'frothing' technique commonly used in 213 the ceramics industry to separate quartz from feldspars in the largest sieve fraction that lacked 214 composite grains (Buckenham and Rogers, 1954). Purified quartz was then used to both measure 215 cosmogenic <sup>3</sup>He abundances and determine sample-specific <sup>3</sup>He diffusion kinetics. All helium 216 isotope measurements were made with an MAP 215-50 sector field mass spectrometer in the 217 BGC Noble Gas Thermochronometry Lab. Gas purification techniques and mass spectrometer 218 analyses are as described in Tremblay et al. (2014b). 219

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Sample	Latitude (decimal degrees)	Longitude (decimal degrees)	Elevation (m)	Sample thickness (cm)	Shielding factor
PDP10	44.421534	7.819950	1806	3	0.9200
PM1	44.252667	7.385500	860	3	0.9653
PM4	44.252667	7.385500	860	3	0.9653
TDB1	44.289712	7.432528	770	3	0.9825
TDB3	44.289712	7.432528	770	3	0.9819

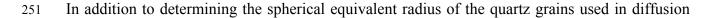
**Table 1:** Locations and sample information for moraine boulders previously dated in the Gesso Valley with cosmogenic <sup>10</sup>Be, originally reported in Federici et al. (2012, 2008).

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For cosmogenic <sup>3</sup>He measurements, 100–500 mg aliquots of quartz were weighed and packed into tantalum metal cups with both ends crimped, placed under vacuum in a sample chamber, and heated with a feedback-controlled 150 W diode laser to either 500, 800, or 1100 °C for 15 minutes until subsequent extractions yielded He signals indistinguishable from the instrumental detection limit. Empty tantalum cup blanks, heated to the same temperatures as the samples, and room temperature procedural blanks were measured throughout each analytical session and subtracted from the sample measurements; <sup>3</sup>He blank corrections were typically 2–3 x  $10^4$  atoms. Aliquots of air and <sup>3</sup>He-spiked helium standards of different manometrically-calibrated pipette volumes were analyzed throughout an instrumental tuning period and used to determine helium sensitivities; sensitivities varied linearly over the pressure range of the sample analyses, estimated by the size of the <sup>4</sup>He signal. We propagated uncertainties from the blank corrections and sensitivity regression into the cosmogenic <sup>3</sup>He concentrations. We assume that all <sup>3</sup>He is cosmogenic.

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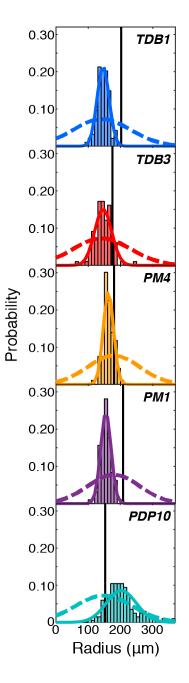
To determine sample-specific diffusion kinetics, we conducted stepwise degassing experiments 236 on a proton-irradiated quartz grain from each sample. Proton irradiation generates a uniform 237 distribution of <sup>3</sup>He in quartz through similar nuclear reactions to those induced by cosmic ray 238 particles but with ten orders of magnitude or higher production rates, enabling step degassing 239 diffusion experiments to be conducted on single irradiated quartz grains (Shuster et al., 2004; 240 Shuster and Farley, 2005; Tremblay et al., 2014b). Quartz aliquots were irradiated with a 228.5 241 MeV proton beam for 6 hours at the Francis H. Burr Proton Therapy Center at the Massachusetts 242 General Hospital in December 2015; the total proton fluence for this irradiation was  $9.14 \times 10^{15}$ 243  $p/cm^2$ . The irradiation target design and setup are detailed in Tremblay et al. (2017). Proton 244 irradiated quartz aliquots were examined with a binocular microscope, and single crystals 245 lacking visible penetrative fractures and inclusions (fluid or mineral) were selected for stepwise 246 degassing. The dimensions of the chosen grains were measured to estimate the spherical 247 equivalent radius used in later calculations to scale the diffusion lengthscale. Details of the 248 stepwise degassing setup and helium measurements are detailed in Tremblay et al. (2014b). 249



experiments, we also determined the spherical equivalent radii of the quartz grains used for 252 cosmogenic measurements and assessed whether these radii are representative of the quartz size 253 distribution in whole rock. Such an assessment is important because if we significantly reduced 254 the grain size of quartz during the crushing process, this could lead to both an underestimation of 255 the diffusion lengthscale to which the diffusion experiment size is scaled and missing 256 cosmogenic <sup>3</sup>He from our measurements. First, we photographed and measured the dimensions 257 of at least 100 quartz grains from the sieve fraction of each sample used for comsogenic <sup>3</sup>He 258 measurements. We determined major, intermediate, and minor axes of best fitting ellipsoids 259 using the software ImageJ (Abràmoff et al., 2004) and calculated the radius of a sphere with the 260 same surface area to volume ratio for each grain. We compared the spherical equivalent radii 261 calculated using this approach to spherical equivalent radii determined from micro x-ray 262 computed tomography (CT) analyses on a smaller number of grains from the same samples, 263 wherein the grains were mapped in three dimensions with a resolution better than 5 micrometers. 264 CT scans were obtained on the Xradia MicroXCT scanner at the University of Texas at Austin 265 High-Resolution X-ray CT Facility and processed using the software Blob3D (Ketcham, 2005) 266 as described in Tremblay et al. (2017). This comparison demonstrates that the ellipsoid approach 267 consistently overestimates the spherical equivalent radius by ~50%, likely because the ellipsoid 268 method significantly underestimates the surface area of grains. We therefore scaled the spherical 269 equivalent radii from loose grain measurements by a factor 1.5; the distribution of grain sizes 270 271 from these measurements are shown in Fig. 2. Also shown in Fig. 2 is the probability distribution of spherical equivalent radii of quartz grains in whole rock. To determine this distribution, we 272 determined circular equivalent radii from measured sectional areas of at least 100 quartz grains 273 274 in thin sections, and inverted the distribution of circular equivalent radii to a distribution of spherical equivalent radii using the code STRIPSTAR and the methodology described by Heilbronner and Barrett (2013). For PDP10, the thin section was made from the same sample as the sample crushed for cosmogenic <sup>3</sup>He measurements. For the PM and TDB moraines, no whole rock material from the original samples was preserved; we therefore collected additional material from boulders exposed atop each of these moraines for thin sections, and assume that the grain size observed in these samples is representative of the samples for which we have cosmogenic <sup>3</sup>He data.

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For all samples, the distribution of spherical equivalent radii for the sieve fraction has a smaller 283 standard deviation than the radii determined from thin section measurements (Fig. 2), which is 284 not surprising given that the sieving process removes larger and smaller grains. With the 285 exception of the PM samples, we find that the mean spherical equivalent radius of quartz in the 286 sieve fraction is equal to or slightly greater than the mean radius determined from thin section 287 measurements (Fig. 2). The mean quartz radius in the sieve fraction from the PM samples is only 288 slightly smaller than the mean radius from thin section measurements, and the two distributions 289 show significant overlap. Given the good agreement between mean spherical equivalent radii for 290 the sieve fraction and thin section quartz, we assume that the grain size has not been substantially 291 reduced by sample crushing and use the mean and standard deviation of the sieve fraction radii 292 distributions as the appropriate diffusion lengthscale for modeling the diffusion of cosmogenic 293 <sup>3</sup>He in each sample. 294



**Figure 2:** Grain size analysis for Gesso Valley samples. Histograms and solid curves denote the distribution of spherical equivalent radii for representative quartz grains ( $\geq 100$ ) from the sieve fraction analyzed for cosmogenic <sup>3</sup>He abundances. Dashed curves denote the distribution of spherical equivalent radii in whole rock, inverted from the distribution of sectional circles of quartz grains ( $\geq 100$ ) measured in thin section. Thin sections were made from the same sample as the crushed fraction, in the case of PDP10, or from a resampled boulder on the same moraine, in all other cases. Vertical black lines denote the spherical equivalent radius of the single quartz grain analyzed in each diffusion experiment shown in Fig. 4.

**3. Results** 

We report measured abundances of cosmogenic <sup>3</sup>He in guartz aliquots from each of the Gesso 298 Valley samples in Table 2. We used version 3 (v3) of the online exposure age calculator code, a 299 more recent version of the code published in Balco et al. (2008) that implements the cosmogenic 300 <sup>3</sup>He production rate in guartz determined by Vermeesch et al. (2009), to calculate an apparent 301 exposure age from the measured <sup>3</sup>He abundance in each aliquot. We assume that the erosion rate 302 is negligible for all boulders sampled, as was assumed in the original publications reporting <sup>10</sup>Be 303 exposure ages from these boulders (Federici et al., 2012, 2008). Sample coordinates, elevations, 304 thicknesses, and shielding correction factors used in exposure age calculations are reported in 305 Table 1. In Table 3, we report the weighted mean apparent <sup>3</sup>He exposure age for each sample 306 determined using the scaling scheme of Stone (2000). We also recalculated the <sup>10</sup>Be exposure 307 ages, normalizing the <sup>10</sup>Be concentrations to the isotope ratio standards of Nishiizumi et al. 308 (2007), using v3 of the online exposure calculator and the global  $^{10}$ Be production rate calibration 309 dataset, and assuming the scaling scheme of Stone (2000) (Table 3). We divided the weighted 310 mean apparent <sup>3</sup>He exposure ages by the <sup>10</sup>Be exposure ages to calculate <sup>3</sup>He retention, which 311 represents the fraction of cosmogenic <sup>3</sup>He produced during surface exposure that remains in the 312 quartz analyzed (Tremblay et al., 2014a). 313

Sample - aliquot	Mass of quartz analyzed (g)	[ <sup>3</sup> He] (10 <sup>3</sup> atoms/g)		
PDP10-1	0.109	2.20	±	0.38
PDP10-2	0.101	2.38	±	0.37
PDP10-3	0.102	3.65	±	0.35
PDP10-4	0.126	2.91	±	0.26
PDP10-5	0.121	2.99	±	0.39
PDP10-6	0.136	2.99	±	0.31

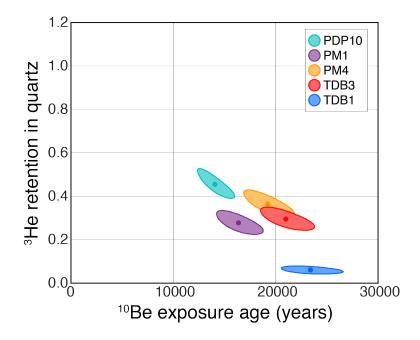
PDP10-7	0.117	3.12	±	0.37
PDP10-8	0.125	2.44	±	0.35
PM1-1	0.101	1.14	±	0.42
PM1-2	0.101	0.89	±	0.6
PM1-3	0.304	1.11	±	0.18
PM1-4	0.266	0.88	±	0.15
PM1-5	0.290	1.05	±	0.15
PM4-1	0.113	1.29	±	0.34
PM4-2	0.309	1.51	±	0.17
PM4-3	0.268	1.63	±	0.19
PM4-4	0.247	1.60	±	0.12
TDB1-1	0.216	0.62	±	0.28
TDB1-2	0.269	0.11	±	0.14
TDB1-3	0.306	0.28	±	0.13
TDB1-4	0.292	0.26	±	0.13
TDB1-5	0.482	0.36	±	0.08
TDB3-1	0.216	1.38	±	0.27
TDB3-2	0.124	1.35	±	0.39
TDB3-3	0.279	1.20	±	0.19
TDB3-4	0.301	1.31	±	0.27
TDB3-5	0.268	1.32	±	0.18

**Table 2:** Observations of cosmogenic <sup>3</sup>He in Gesso Valley quartz samples.

Fig. 3 shows <sup>3</sup>He retention as a function of exposure duration in the five samples we analyzed. All Gesso Valley samples retain less than 50% of the cosmogenic <sup>3</sup>He that was produced during their exposure (Table 3, Fig. 3). Retention generally decreases with increasing exposure

duration: guartz from PDP10, sampled from the highest elevation moraine with an exposure age 319 of  $14071 \pm 1220$  years, retains 46% of its cosmogenic <sup>3</sup>He, while guartz from TDB1, sampled 320 from the lowest elevation moraine with an exposure age of  $23397 \pm 2072$  years, retains only 6% 321 of its cosmogenic <sup>3</sup>He. Samples PM1 and PM4, both from the Ponte Murato moraine, have <sup>10</sup>Be 322 exposure ages that agree within uncertainty  $(16356 \pm 1601)$  years and  $19213 \pm 1654$  years, 323 respectively) but significantly different <sup>3</sup>He retention  $(0.28 \pm 0.04 \text{ versus } 0.36 \pm 0.04,$ 324 respectively; Table 3, Fig. 3). Similarly, the <sup>10</sup>Be exposure ages for the two Tetti del Bandito 325 moraine samples, TDB1 and TDB3, are in agreement (23397  $\pm$  2072 years and 21003  $\pm$  1854 326 years, respectively), but the <sup>3</sup>He retentions in these two samples are drastically different (0.06  $\pm$ 327 0.01 versus  $0.29 \pm 0.04$ , respectivley; Table 3, Fig. 3). 328

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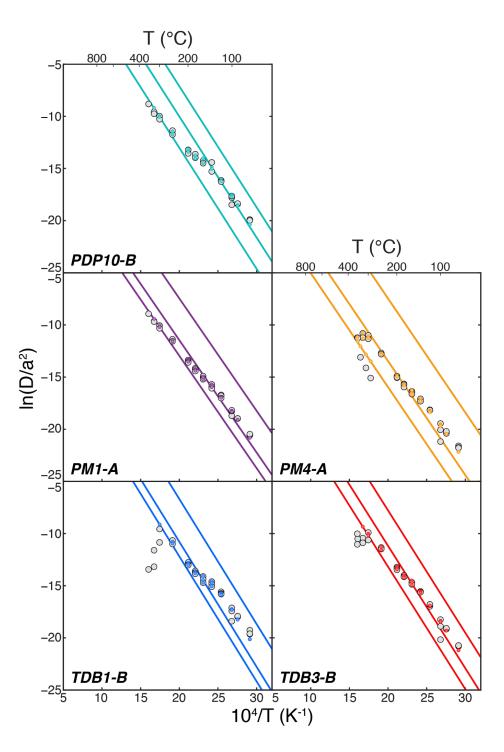
**Figure 3:** Cosmogenic <sup>3</sup>He retention as a function of <sup>10</sup>Be exposure age in quartz from Gesso Valley moraine samples. Ellipses represent  $1\sigma$  uncertainty.

Sample	Apparent exposure age (yr), <sup>3</sup> He			Exposure age (yr), <sup>10</sup> Be			<sup>3</sup> He retention		
PDP10	6402	±	341	14071	±	1220	0.46	±	0.05
PM1	4534	±	452	16356	±	1601	0.28	±	0.04
PM4	7006	±	512	19213	±	1654	0.36	±	0.04
TDB1	1407	±	271	23397	±	2072	0.06	±	0.01
TDB3	6187	±	561	21003	±	1854	0.29	±	0.04

**Table 3:** Apparent <sup>3</sup>He exposure ages and retention in Gesso Valley quartz samples. See text for details about the input parameters to the exposure age calculation. We report external uncertainties for both <sup>3</sup>He and <sup>10</sup>Be exposure ages.

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In order to reconstruct effective diffusion temperatures (EDTs), and by relation temperature histories, from the observed <sup>3</sup>He retention in quartz from each sample and assess the differences in <sup>3</sup>He retention between samples from the same moraine, we need to know the diffusion kinetics of <sup>3</sup>He in each sample. Fig. 4 shows the results of step degassing experiments on protonirradiated quartz from the Gesso Valley samples in Arrhenius plots where the natural log of diffusivity, calculated from the cumulative fraction of gas released during each heating step (Fechtig and Kalbitzer, 1966), is plotted as a function of inverse temperature (gray circles).



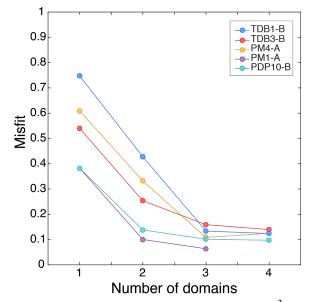
**Figure 4:** Arrhenius plots for helium diffusion experiments on Gesso Valley quartz samples.  $D/a^2$  values are normalized to  $s^{-1}$ . Experimental data are plotted as gray circles. Diffusivities were calculated using the equations of Fechtig and Kalbitzer (1966) and the uncertainty propagation outlined in Tremblay et al. (2014b). Color circles and lines denote three-domain multiple diffusion domain (MDD) models that minimize misfit with the experimental data. Tremblay et al. (2017) describe MDD modeling approach used here.

340 All Gesso Valley quartz samples exhibit some degree of nonlinearity in the Arrhenius plots (Fig. 4). While we do not have a mechanistic explanation or model for this type of complex diffusion 341 behavior (Tremblay et al., 2014b), multiple diffusion domain (MDD) models (Harrison et al., 342 1991; Lovera et al., 1991; Lovera and Richter, 1989) can reproduce the nonlinearity observed in 343 laboratory degassing experiments and appear to be relevant to <sup>3</sup>He diffusive loss over  $10^{3}$ – $10^{4}$ 344 year timescales, as demonstrated for a geologic case study for which the temperatures during 345 surface exposure are reasonably well known (Tremblay et al., 2014b). We constructed MDD 346 models optimized to fit each of the helium step degassing experiments on Gesso Valley quartz 347 samples following the approach outlined in Tremblay et al. (2017). Fig. 5 shows the misfit 348 between the calculated diffusivities from MDD models and the experimentally-determined 349 diffusivities as a function of number of diffusion domains, where the misfit statistic was 350 calculated as in Tremblay et al. (2017). The misfit shown for a given number of diffusion 351 domains is the minimum misfit found after searching over a large range of model parameters 352 (activation energy  $E_a$ , pre-exponential factor(s)  $D_0/a^2$ , and gas fraction(s)). 353

354

We found that, for all five step degassing experiments, increasing the number of diffusion 355 domains from 1 to 2 and from 2 to 3 significantly reduced the misfit between the observed 356 diffusivities and those calculated with the MDD model (Fig. 5). However, increasing the number 357 of diffusion domains from 3 to 4 did not significantly reduce the misfit between the observed and 358 359 calculated diffusivities (Fig. 5). For experiment PM1-A, attempts at constructing a four-domain model yielded two domains with the same pre-exponential factor and thus are the equivalent of 360 the three-domain model. Therefore for all experiments we use the diffusion parameters 361 362 corresponding to the optimized three domain model. Lines corresponding to the diffusion

parameters of the three domains, as well as the calculated diffusivities for the three domain models, are shown in color over the experimentally determined diffusivities in Fig. 4. The activation energies, pre-exponential factors, gas fractions, and misfit statistic for these three domain models are reported in Table 4.



**Figure 5:** Minimized misfit between observed and MDD-modeled <sup>3</sup>He diffusivities as a function of number of diffusion domains for Gesso Valley quartz samples. We increased the number of diffusion domains in our MDD models until the addition of another diffusion domain did not significantly reduce the misfit between the observed and calculated diffusivities. For all Gesso Valley quartz samples, we found that increasing the number of domains from three to four only marginally improved the misfit; therefore we use three-domain MDD models in subsequent calculations. For experiment PM1-A, attempts at constructing a four domain model yielded two domains with the same pre-exponential factor and thus are the equivalent of the three domain model.

Experiment	Domain	E <sub>a</sub> (kJ/mol)	$\ln(D_0/a^2)$	fraction	Misfit
PDP10-B	1	96.5	10.2	0.61	0.097
	2		16.1	0.21	
	3		13.2	0.18	
PM1-A	1	89.6	8.6	0.54	0.067
	2		10.1	0.36	
	3		14.1	0.1	

PM4-A	1	90.9	8.4	0.74	0.109
	2		5.9	0.2	
	3		14.4	0.06	
TDB1-B	1	99.7	11.8	0.46	0.161
	2		13.2	0.3	
	3		17.3	0.24	
TDB3-B	1	97.6	10.3	0.42	0.149
	2		12.3	0.41	
	3		15.7	0.17	

**Table 4:** Three domain multiple diffusion domain model parameters for <sup>3</sup>He diffusion experiments in Gesso Valley quartz samples.

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As discussed in Section 2, we need to scale the diffusion kinetics determined from the step 369 degassing experiments to the diffusion lengthscale appropriate for the cosmogenic <sup>3</sup>He 370 abundance measurements. In constructing MDD models, we cannot independently parameterize 371 the diffusivity at infinite temperature,  $D_0$ , and the diffusion lengthscale, a. Instead, we model 372 373 them together as the natural logarithm of the pre-exponential factor in the Arrhenius expression,  $ln(D_0/a^2)$ , which in an Arrhenius plot corresponds to the y-intercept. Because we cannot 374 independently model the diffusion lengthscale a, we must make an assumption about how to 375 scale the MDD models appropriately. This is not a problem for the  ${}^{40}$ Ar/ ${}^{39}$ Ar thermochronometry 376 community in which MDD models were developed, because in almost all cases argon diffusion 377 kinetics and naturally-occurring argon isotope abundances are measured in the same mineral 378 grain (e.g. Lovera et al., 1997/8). Here, we assume that the diffusion lengthscale of all domains, 379 and by inference all the pre-exponential factors in a given MDD model, scales with the spherical 380 equivalent radius of the quartz grain analyzed. Theoretically, this implies that diffusion 381 lengthscale of the most retentive domain (the domain with the lowest  $ln(D_0/a^2)$  or intercept in 382 Arrhenius space) is the spherical equivalent radius of the grain analyzed. Practically, this scaling 383

is accomplished as follows:

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386 
$$\left(\frac{D_0}{a^2}\right)_{scaled} = \left(\frac{D_0}{a^2}\right)_{MDD} \left(\frac{a_{sde}^2}{a_{cosmo}^2}\right)$$
 (1)

387

where  $a_{sde}$  is the spherical equivalent radius of the step degassing experiment quartz grain and *a<sub>cosmo</sub>* is the spherical equivalent radius of the quartz grains used for the cosmogenic <sup>3</sup>He measurements (Fig. 2).

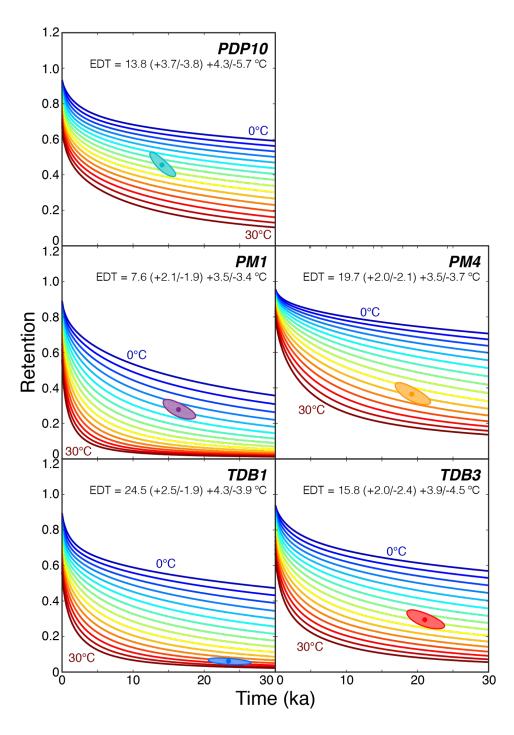
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With the scaled, sample-specific diffusion kinetics, we explore possible temperature scenarios to 392 explain the observed cosmogenic <sup>3</sup>He abundances in the Gesso Valley samples. To do this, we 393 use forward models of simultaneous production and diffusion to model how cosmogenic <sup>3</sup>He 394 retention evolves as a function of exposure duration under different temperature scenarios. We 395 begin with the simplest possible scenario of constant temperature over time, as this allows us to 396 assess the EDT integrated over the exposure duration of each sample. These EDTs are equal to 397 or greater than the mean temperatures the samples experienced over their exposure durations 398 because of the nonlinear relationship between diffusivity and temperature (Tremblay et al., 399 2014a). Fig. 6 shows the evolution of <sup>3</sup>He retention as a function of exposure duration for 400 401 constant temperature scenarios in 2 °C increments from 0 °C to 30 °C. These retention evolution curves differ for each Gesso Valley quartz sample for several reasons: (1) each sample has a 402 403 different set of diffusion kinetics, assuming a three domain diffusion model (Fig. 4, Table 4), (2) 404 these sample specific diffusion kinetics are scaled to different spherical equivalent radii based on the measured grain size distributions of quartz in the sieve fraction used for cosmogenic <sup>3</sup>He 405 measurements (Fig. 2), and (3) the cosmogenic <sup>3</sup>He production rate in quartz differs between 406

samples. In each panel of Fig. 6 we report the integrated EDT that agrees with the observed cosmogenic <sup>3</sup>He retention in each quartz sample. We provide two uncertainty estimates on this integrated EDT. One  $\sigma$  EDT uncertainties in parentheses account for the uncertainty in <sup>3</sup>He retention and an exposure duration as determined from <sup>10</sup>Be measurements; the second set of uncertainties also account for uncertainty in the spherical equivalent radius to which the experimentally-determined diffusion kinetics are scaled.

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The most readily observable feature of the models in Fig. 6 is that the integrated EDT calculated 414 for each quartz sample is different. This is in part expected because the different moraines 415 represented by these samples are located at different elevations in the Gesso Valley, and thus will 416 experience different temperatures at any point in time due to the atmospheric lapse rate. This 417 may also in part be due to the samples having different exposure durations; for example, a 418 change in temperature between the deposition of the TDB moraine and the PM moraine would 419 only be reflected in the cosmogenic <sup>3</sup>He retention and integrated EDT of the TDB moraine 420 samples. Differences in integrated EDT between samples from the same moraine, on the other 421 hand, are unexpected. For PM1 we calculate an integrated EDT of 7.6 °C, while for PM4 we 422 calculate a much higher integrated EDT of 19.7 °C. Similarly, for TDB1 we calculate an 423 integrated EDT of 24.5 °C, while for TDB3 we calculate a much lower integrated EDT of 15.8 424 °C. 425

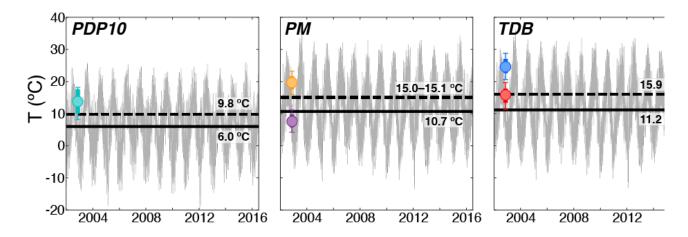


**Figure 6:** EDTs integrated over the exposure duration of Gesso Valley moraine samples. For each sample, we plot cosmogenic <sup>3</sup>He retention as a function of exposure duration assuming a constant EDT and the MDD model diffusion kinetics reported in Table 3. Observed cosmogenic <sup>3</sup>He retention ellipses are plotted as in Fig. 4 for comparison. One  $\sigma$  EDT uncertainties in parentheses account for the uncertainty in <sup>3</sup>He retention and exposure duration as determined from <sup>10</sup>Be measurements; the second set of uncertainties also account for uncertainty in the spherical equivalent radius to which the experimentally-determined diffusion kinetics are scaled.

To put these integrated EDTs into context, in Fig. 7 we use modern daily temperature records 427 from 2002 to 2016 to calculate mean temperatures (solid lines) and EDTs (dashed lines) at the 428 elevation of each moraine. We obtained temperature records at the elevation of each moraine by 429 scaling daily mean, minimum, and maximum temperature data from two meteorological stations 430 within the Gesso Valley: Diga della Piastra (44.227 °E, 7.389 °N, 959 m) and Diga del Chiotas 431 (44.168 °E, 7.334 °N, 1980 m). These data are publicly available through the Environmental 432 Protection Agency for the Piemonte Region (arpa.piemonte.gov.it). We scaled the temperature 433 data at Diga della Piastra to the elevation of the TDB and PM moraines and the temperature data 434 at Diga del Chiotas to the elevation of the PDP moraine using monthly average lapse rates 435 determined from the same dataset, and calculated the modern EDT using these scaled 436 temperature data and the activation energies of the best fit MDD models in Fig. 4 and Table 4. 437 For PDP10, PM4, and TDB1, the modern EDT estimate (9.8, 15.1, and 15.9 °C) is several 438 degrees lower than the EDT integrated over each sample's exposure duration (13.8, 19.7, and 439 24.5 °C). In contrast, the modern EDT estimate for TDB3 (15.9 °C) is comparable to the EDT 440 integrated over its exposure duration (15.8 °C), and for PM1 the modern EDT estimate (15.0 °C) 441 is several degrees higher (vs. 7.6 °C). 442

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In addition to comparing the integrated EDTs calculated from observed cosmogenic <sup>3</sup>He abundances to modern day EDTs in the Gesso Valley, we can compare our cosmogenic <sup>3</sup>He observations to the those that would be result (1) from temperature histories simulated in general circulation models (GCMs), and (2) from temperature reconstructions based on other proxies at nearby locations. For both model and proxy comparisons, the temperature history used to model cosmogenic <sup>3</sup>He abundances must be equal in duration to the moraine sample exposure.



**Figure 7:** EDTs calculated from modern meteorological data. In gray we show the time series of mean, minimum, and maximum daily temperatures from 2002 to mid-2016 at the elevations of the PDP, PM, and TDB moraines, scaled from the nearest two meteorological stations using average monthly lapse rates. Solid black bars denote the mean temperature for each time series; dashed black bars denote the modern EDT for each time series calculated using the activation energies of the best fit MDD models in Fig. 4 and Table 4. EDTs integrated over the exposure duration of each sample are also shown for comparison as circles, with colors for specific samples and  $1\sigma$  uncertainties as reported in Fig. 6.

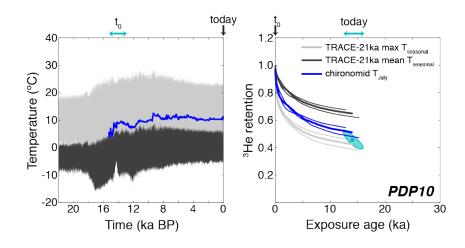
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Most GCM paleo-experiments simulate climate at a particular point in time, such as the mid-451 Holocene or LGM; only a handful of GCMs simulations have explored transient climate 452 evolution in the Holocene and latest Pleistocene. Here, we model how cosmogenic <sup>3</sup>He retention 453 would evolve as a function of exposure duration using seasonal temperature outputs from the 454 TraCE-21ka transient simulation. TraCE-21ka uses the National Center for Atmospheric 455 Research (NCAR) Community Climate System Model version 3 (CCSM3), a synchronously 456 coupled atmosphere-ocean general circulation model (GCM) (Collins et al., 2006), to simulate 457 458 the Earth's climate from the LGM (22 thousand years ago) to the present (He, 2011; Liu et al., 2009). We obtained seasonal (DJF, MAM, JJA, and SON) mean and maximum temperatures 459 from the TraCE-21ka simulation for the 44.3 °N, 7.4 °E grid cell including the Gesso Valley. 460 461 These simulated temperatures were determined for a mean elevation above sea level for the grid

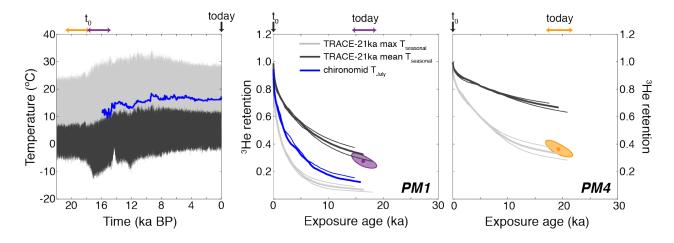
cell, which changed through the simulation as simulated sea level changed. However, the 462 elevation of our sample sites with respect to sea level would have also changed through time, 463 meaning that the relative offset between our sample site elevation and the mean grid cell 464 elevation would have remained constant. We therefore scale the TraCE-21ka simulation 465 temperatures to our sample sites using the present day difference between the sample site 466 elevations and TraCE-21ka mean elevation assuming the modern seasonal lapse rate determined 467 from the meteorological stations discussed above. The atmospheric lapse rate in the Gesso 468 Valley likely changed between the LGM and the present, especially considering the fact that the 469 valley was more extensively glaciated during part of this interval. Nonetheless, we assume the 470 modern rate because (1) we have no independent means for assessing how different paleo-lapse 471 rates may have been, and (2) any differences from the modern lapse rate when the valley was 472 glaciated were likely  $\leq 2$  °C/km (e.g. Davis et al., 2003; Gardner et al., 2009; Loomis et al., 473 2017), which will not be resolvable using our cosmogenic noble gas observations that span  $\leq 1$ 474 km in elevation (Tremblay et al., 2014a). 475

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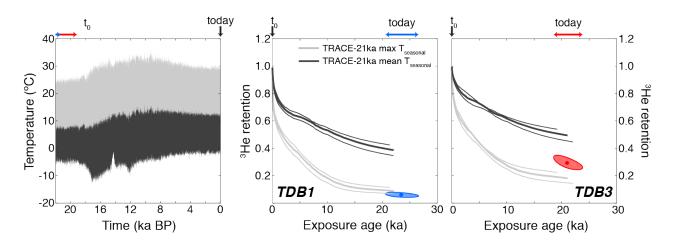
For all Gesso Valley quartz samples, the observed cosmogenic <sup>3</sup>He retention is bracketed by the 477 <sup>3</sup>He retention calculated assuming mean seasonal temperatures and the <sup>3</sup>He retention calculated 478 assuming maximum seasonal temperatures scaled from the TraCE-21ka simulation. For PDP10 479 (Fig. 8), PM4 (Fig. 9), and TDB1 (Fig. 10), we calculate cosmogenic <sup>3</sup>He retention from the 480 TraCE-21ka simulation maximum seasonal temperature that is in agreement within uncertainty 481 with the observed retention in these samples. Cosmogenic <sup>3</sup>He retention calculated from the 482 TraCE-21ka mean seasonal temperatures agrees with the observed cosmogenic <sup>3</sup>He retention in 483 PM1 within uncertainty (Fig. 9), while for TDB3 the observed cosmogenic <sup>3</sup>He retention lies 484



**Figure 8:** Cosmogenic <sup>3</sup>He retention in PDP10 quartz calculated from scaled TraCE-21ka seasonal temperatures and scaled, spliced chironomid July temperatures. The left panel shows the mean (black) and maximum (gray) seasonal temperature from the TraCE-21ka climate simulation and the mean July temperature (blue) from four spliced chironomid records, scaled to the elevation of PDP10 using modern lapse rate data and, for the chironomid records, weighted by distance from our study site. The right panel shows the evolution of <sup>3</sup>He retention with time assuming the TraCE-21ka and chironomid temperatures represent the EDT experience over PDP10's exposure duration. Thick retention curves assume the mean exposure duration and grain size, while upper thin curves assume the  $-1\sigma$  exposure duration and grain size. The ellipse shows the observed retention and exposure duration, as in Fig. 3.



**Figure 9:** Cosmogenic <sup>3</sup>He retention in PM1 and PM4 calculated from scaled TraCE-21ka seasonal temperatures and, in the case of PM1, scaled, spliced chironomid July temperatures. The left panel shows the mean (black) and maximum (gray) seasonal temperature from the TraCE-21ka climate simulation and the mean July temperature (blue) from four spliced chironomid records, scaled to the elevation of PM using modern lapse rate data and, for the chironomid records, weighted by distance from our study site. The right panels show the evolution of <sup>3</sup>He retention with time assuming the TraCE-21ka and chironomid temperatures represent the EDT experience over the PM samples' exposure duration. Thick and thin retention curves are as in Fig. 8. Ellipses show the observed retention and exposure duration, as in Fig. 3.



**Figure 10:** Cosmogenic <sup>3</sup>He retention calculated in TDB1 and TDB3 from scaled TraCE-21ka seasonal temperatures. The left panel shows the mean and maximum seasonal temperature from the TraCE-21ka climate simulation, scaled to the elevation of the TDB moraine using modern lapse rate data. The right panels shows the evolution of <sup>3</sup>He retention with time assuming the TraCE-21ka temperatures represent the EDT experience over the TDB samples' exposure duration. Thick and thin retention curves are as in Fig. 8. Ellipses show the observed retention and exposure duration, as in Fig. 3.

We also use chironomid-based July temperature reconstructions from other Alpine locations to 491 model cosmogenic <sup>3</sup>He retention for comparison with our cosmogenic <sup>3</sup>He observations. The 492 lack of Alpine chironomid records before 16 cal. years BP prevents us from using chironomid-493 based July temperature reconstructions to model cosmogenic <sup>3</sup>He retention in PM4, TDB1, and 494 TDB3. However, we can splice together chironomid records from several locations to obtain 495 coeval coverage with samples PDP10 and PM1, which have the youngest <sup>10</sup>Be exposure ages of 496  $14071 \pm 1220$  and  $16356 \pm 1601$  years, respectively. The most proximal chironomid-based mean 497 498 July temperature reconstruction is from Lago Piccolo di Avigliana (LPA), located in the Italian Alps ~80 km north of the Gesso Valley (45.05 °N; 7.38 °E, 365 m), where the record extends 499 from 14200 to 9500 cal. year BP (Larocque and Finsinger, 2008). In order to obtain coeval 500 coverage with the exposure ages of PDP10 and PM1, we spliced the LPA record with 501 chironomid July temperature reconstructions from: Schwarzsee ob Sölden (SOS), a high-alpine 502 lake in the Austrian Alps (46.96583 °N, 10.94611 °E, 2796 m) with a chironomid record from 503 10200 to -36 cal. years BP (Ilyashuk et al., 2011); Lac Lautrey (LAU), a small lake in the French 504 Jura Mountains (46.58722 °N, 5.86389 °E, 788 m) with a chironomid record from 15908 to 505 11033 cal. years BP (Heiri and Millet, 2005); and Hinterburgsee (HIN), a subalpine lake in the 506 northern Swiss Alps (46.71833 °N, 8.06750 °E, 1515 m) with a chironomid record from from 507 12210 to 1900 cal. years BP (Heiri et al., 2004). Like the GCM simulation, we scaled the July 508 509 temperature reconstructions from each location to the elevations of PDP10 and PM1 using modern lapse rate information, and when temporal overlap occurred between two or more 510 chironomid records we used an average of the scaled temperatures weighted by the inverse 511 512 distance between the chironomid sites and our study site. In Figs. 8 and 9 we show the scaled,

spliced July temperature reconstruction using all four chironomid records and the corresponding 513 model for cosmogenic <sup>3</sup>He retention in PDP10 and PM1, respectively. We found that different 514 chironomid record splices (e.g., SOS + LPA; SOS + HIN + LPA; SOS + LPA + LAU; SOS + 515 HIN + LPA +LAU) result in very small differences in modeled cosmogenic <sup>3</sup>He retention; 516 therefore only the four record splice (SOS + HIN + LPA +LAU) is shown. Like the maximum 517 seasonal temperatures from the TraCE-21ka simulation, we model cosmogenic <sup>3</sup>He retention 518 from the mean July temperature reconstruction that agrees with our observations from PDP10 519 and that underestimates the observed <sup>3</sup>He retention in PM1. 520

521

### 522 4. Discussion

To a first order, the fact that modern EDTs calculated from meteorological data are within a few 523 degrees of the EDTs integrated over the exposure duration of each Gesso Valley quartz sample, 524 and the fact that cosmogenic <sup>3</sup>He retention calculated with temperatures from a GCM simulation 525 and an independent proxy reconstruction are in broad agreement with our cosmogenic <sup>3</sup>He 526 observations, is promising. This first order agreement suggests that our cosmogenic <sup>3</sup>He 527 measurements are, in fact, providing information on ambient temperature conditions during 528 exposure. For example, we do not calculate integrated EDTs below 0 °C, as we do for 529 cosmogenic <sup>3</sup>He measurements made on Holocene-age glacial erratics in Antarctica (Tremblay et 530 al., 2014a), nor do we calculate integrated EDTs unfeasibly high for Earth surface conditions 531 (e.g. EDTs of 70-80 °C that we obtain for the Moon (Shuster and Cassata, 2015). The apparent 532 agreement of some of our cosmogenic <sup>3</sup>He observations with maximum seasonal temperatures 533 from the TraCE-21ka simulation, which has a 3.75° resolution, and with July temperatures 534 535 reconstructed from Alpine chironomid records spread over hundreds of kilometers, also suggests

that the integrated temperature history of the Maritime Alps since the LGM may not have been exceptionally different from those in Alpine regions further north and further inland. However, for the purposes of reconstructing past climate variations, the substantial intra-moraine differences in EDTs are problematic and require explanation. For both the PM and TDB moraines, the difference in integrated EDTs between samples from the same moraine is ~10 °C.

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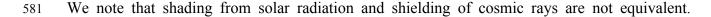
One possibility is that these intra-moraine temperature differences are real and reflect differential 542 shading of the samples by vegetation, snow cover or topography. For example, patchy vegetation 543 on a moraine crest may cause one boulder to be fully shaded under a tree canopy, while another 544 boulder tens of meters away remains unshaded. Similarly, boulders nearby to one another might 545 accumulate substantial or insignificant snow cover depending on their relative positioning and 546 shielding from prevailing winds during the wintertime. Unshaded rock samples can experience 547 significantly higher daytime temperatures due to insolation than shaded samples. As discussed in 548 Tremblay et al. (2014a), heating of rocks due to incident solar radiation can substantially raise 549 rock temperatures above daily maximum air temperatures. Amplification of temperatures 5-10550 °C above the daily maximum is common in non-desert environments (Bartlett et al., 2006; Hall et 551 al., 2005; McGreevy, 1985; Schwarz et al., 2012). Considering our modern EDT calculations 552 from meteorological station data, if we increase the maximum daily temperature by 5 or 10 °C, 553 we find significant increases in the EDT. For example, using the diffusion kinetics for TDB1, by 554 555 increasing the maximum daily temperature in the meteorological data by 5 °C, we find that EDT increases from 15.9 to 19.2 °C. Increasing the maximum daily temperature by 10 °C increases the 556 EDT to 23.0 °C, which is within the uncertainty of the EDT integrated over TDB1's exposure 557 558 duration. Therefore it is possible that, in the case of the TDB moraine, TDB3 may have been

shaded or snow covered during all or some of its exposure history, while TDB1 was fully
exposed to incoming solar radiation throughout its exposure. Similarly, PM1 may have been
shaded or snow covered while PM4 was not since the PM moraine was deposited.

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This interpretation is consistent with the comparison of observed cosmogenic <sup>3</sup>He retention to 563 calculated retention from TraCE-21ka and chironomid temperatures. TraCE-21ka simulation 564 maximum seasonal temperature calculations agree well with PDP10, PM4, and TDB1 565 observations, while observations from PM1 and TDB3 are closer to the calculations with the 566 mean seasonal temperature from TraCE-21ka. We would expect mean temperatures to agree 567 with our cosmogenic <sup>3</sup>He observations, as is the case for PM1, if no heating due to insolation 568 occurred. Following the same reasoning, we anticipate that samples experiencing nonzero 569 insolation will have cosmogenic <sup>3</sup>He abundances that are lower than what is expected from mean 570 air temperatures, as is the case for PDP10, PM4, TDB1, and TDB3. Likewise, the good 571 agreement between the observed cosmogenic <sup>3</sup>He retention in PDP10 and the chironomid-based 572 reconstruction of mean July temperatures, which will be significantly greater than mean 573 temperatures throughout most of the year, supports the interpretation that the temperatures 574 experienced by sample PDP10 exceeded air temperatures during its exposure to cosmic ray 575 particles. Relating maximum seasonal air temperatures or mean July air temperatures to 576 insolation-induced daily rock temperature amplifications is not straightforward, although an 577 578 empirical relationship could be obtained by monitoring *in situ* rock temperatures adjacent to meteorological stations. 579

580



Vegetative cover can have a substantial effect on heating by insolation, but in most cases a 582 negligible effect on cosmogenic nuclide production rates due to its relatively low density. 583 Similarly, thin snow cover will not substantially affect cosmogenic nuclide production rates but 584 will cause rock temperatures to differ substantially from air temperatures. Thick snow cover ( $\geq 1$ 585 m) will affect both production rates and temperatures. Today, there is significant vegetation 586 587 cover on the TDB and PM moraines, but no cover at PDP. Unfortunately, we have no way of assessing the degree of vegetation cover on the TDB and PM moraines before the modern, and 588 no way to assess the past degree of snow cover for all moraines. Topography can shield rocks 589 590 from cosmic ray particles, but these effects are accounted for in our calculations of cosmogenic nuclide production rates (Table 1); in contrast, we have not accounted for the effects of 591 topographic shading on rock surface temperatures. We do not have detailed enough locations for 592 the TDB and PM samples to identify the original boulders that were sampled and assess 593 differences in topographic shading, although we suspect this would be secondary to vegetation 594 and snow cover effects. 595

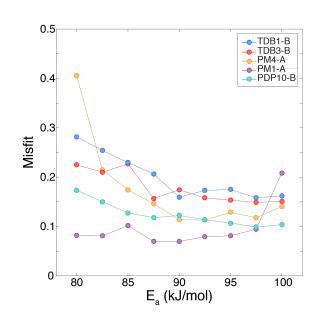
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Another possible explanation for the intra-moraine differences is that our MDD model 597 598 representations of helium diffusion kinetics in Gesso Valley quartz samples are inaccurate and/or inadequate. The MDD models we construct are inherently non-unique, and while the models we 599 use minimize the misfit between the observed and calculated diffusivities in our step degassing 600 601 experiments for a given number of domains, comparable fits can be achieved over a range of diffusion parameters. To illustrate this, in Fig. 11 we show minimized values of MDD model 602 misfit, calculated using the misfit statistic defined in Tremblay et al. (2017), as a function of 603 604 activation energy  $E_a$  for three domain models fit to each of the step degassing experiments

shown in Fig. 4. For most of the step degassing experiments, there is a broad range of activation energies (and associated pre-exponential factors and gas release fractions) over which the misfit with the MDD model changes only slightly; these changes in misfit are much smaller than the change in misfit associated with increasing the number of diffusion domains and the model complexity (Fig. 5).

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**Figure 11:** Minimized misfit between observed and MDD-modeled <sup>3</sup>He diffusivities as a function of activation energy  $E_a$  for Gesso Valley quartz samples, assuming a three-domain diffusion model. Pre-exponential factors and gas fractions for each domain covary with  $E_a$ ; we show misfit as a function of  $E_a$  because it is assumed to be common to all diffusion domains in our modeling approach.

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For example, the MDD misfit for the PM1-A step degassing experiment ranges between 7.0 and 10.2% between activation energies of 80 and 97.5 kJ/mol (Fig. 11). The MDD diffusion kinetics we use in the forward models for PM1 quartz presented above assume an activation energy of 89.6 kJ/mol. If instead we use a different set of diffusion kinetics for a three-domain model that

yields a comparable misfit with the step degassing experiment ( $E_a = 97.5 \text{ kJ/mol}$ ;  $ln(D_0/a^2) =$ 617 12.2, 10.1, 16.8;  $f_{gas} = 0.45$ , 0.44, 0.11; Misfit = 0.095), we calculate an integrated EDT from the 618 cosmogenic <sup>3</sup>He retention in PM1 quartz of 17.8 ( $\pm$  2.3) +3.8/-3.9 °C, which is in agreement with 619 the integrated EDT we calculated for PM4 (Fig. 6). This highlights how sensitive the 620 temperatures we calculate from an observed amount of cosmogenic <sup>3</sup>He retention are to the 621 diffusion kinetics we use. While different MDD models might reproduce diffusivities observed 622 in laboratory step degassing experiments comparably well, the downward extrapolation of these 623 models results in significantly different diffusivities at the temperatures characterizing Earth's 624 surface. This suggests that some or all of the intra-moraine differences in integrated EDTs could 625 be attributed to uncertainties in how we extrapolate laboratory-determined diffusion kinetics. 626 Given that we do not yet have a mechanistic understanding of what controls the complex noble 627 gas diffusion behavior we observe in our experiments, discriminating between different MDD 628 models of comparably good fit (and even assessing whether MDD models adequately represent 629 the processes responsible for complex behavior) is difficult at this stage. 630

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A third possible explanation for the intra-moraine variation could arise from our grain size 632 analysis and scaling of MDD models. Uncertainty in the physical significance of our MDD 633 models translates into an uncertainty in our assumptions about how to scale our MDD model fits 634 from step degassing experiments to the cosmogenic <sup>3</sup>He measurements. However, because we 635 636 have scaled the results of each experiment using the same assumptions in Eq. 1, the effects will be systematic. Similarly, the factor of 1.5 we applied to estimated spherical equivalent radii of 637 quartz from the sieve fractions, based on different approaches to estimate surface area to volume 638 639 ratios and ground truthing with x-ray computed tomography data, will have systematic effects on

all integrated EDTs. Assuming that the MDD model scaling and correction applied to sieve 640 fraction measurements are appropriate, we must also consider the possibility that our grain size 641 analysis for the PM and TDB samples does not represent the actual grain size and diffusion 642 lengthscale of quartz in these samples. Since we did not have whole rock material from the 643 original PM and TDB samples, we made thin sections from other boulders collected from the PM 644 and TDB moraines to compare with the size distributions of quartz in the sieved fraction. While 645 there is good agreement between the mean spherical equivalent radii of quartz measured in these 646 thin sections and in the sieve fractions, it is possible that in the original sample the mean quartz 647 size before crushing was much larger. As a sensitivity test, we again use PM1 as an example. In 648 order to obtain an integrated EDT for PM1 equivalent to that calculated for PM4 of 19.7 °C, we 649 must increase the spherical equivalent radius assumed for the cosmogenic <sup>3</sup>He measurement by 650 110% (without changing the spherical equivalent radius for PM4). While such a dramatic 651 underestimation of the PM1 grain size, and more generally >100% uncertainty in all our PM and 652 TDB grain size analyses, seems unlikely, we cannot rule out such a possibility given that whole 653 rock material from these samples was unavailable. 654

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In summary, there is substantial intra-moraine variability in the integrated EDTs we calculate from our cosmogenic <sup>3</sup>He observations that requires explanation. Nonetheless, we calculate an integrated EDT for at least one sample from each moraine that is consistent with maximum seasonal temperatures in the TraCE-21ka simulation and from chironomid-based mean July temperature reconstructions. Given that we expect integrated EDTs to exceed the mean ambient temperatures in a sample's exposure history because of the nonlinear dependence of diffusivity on temperature, this agreement is promising for the application of cosmogenic noble gas

paleothermometry in paleoclimate studies. For the Maritime Alps specifically, this result may 663 indicate that this region experienced a Late Quaternary climatic history similar to the rest of the 664 western Alps, and that proximity to the Mediterranean Sea had a limited effect on the 665 temperatures of this region. Future application of cosmogenic noble gas paleothermometry to a 666 larger number of samples per moraine, as well as to more recently formed landforms whose 667 shading history can be better resolved, accompanied with further advances in our understanding 668 of noble gas diffusion kinetics, will allow for much improved reconstruction of 669 paleotemperatures in the future. 670

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## 672 **5. Conclusions**

In summary, integrated EDTs since the LGM, calculated from our observations of cosmogenic 673 <sup>3</sup>He concentrations in quartz range from 8 to 25 °C and are broadly consistent with what we 674 expect from modern meteorological station data, a GCM simulation of Earth's climate since the 675 LGM, and data from independent proxies in the region. Nonetheless, there are nontrivial 676 differences between the integrated EDTs we calculate, particularly for samples with equivalent 677 exposure ages from the same moraine, that require explanation. We identify three major potential 678 sources of uncertainty that could account for these discrepancies: (1) the fact that we have not 679 accounted for variations in radiative heating of the boulder surfaces across samples due to 680 differential shading, (2) limitations in our understanding of and ability to model and extrapolate 681 682 helium diffusion kinetics when complex behavior is observed, or (3) uncertainties associated with our quartz grain size analyses. At this stage, all three of these possibilities could be 683 contributing to intra-moraine and inter-moraine variability in reconstructed temperatures. 684 Because of these nontrivial uncertainties, and the fact that we have no cosmogenic <sup>3</sup>He 685

observations from younger moraines that record only more recent, Holocene temperatures in the Gesso Valley, it is difficult to fully compare our results to the TraCE-21ka and chironomid temperature records and not yet possible to comment on the hypothesis that the post-LGM climate evolution and glacier dynamics of the Maritime Alps differed from other Alpine regions. Nonetheless these results are promising, and suggest that with additional work and better understanding of diffusion kinetics and rock-air temperature relationships we can address questions in paleoclimate like this one in future applications.

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