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1 Anomalously old biotite $^{40}\text{Ar}/^{39}\text{Ar}$ ages in the NW Himalaya

2
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12 13 **ABSTRACT**

14 Biotite $^{40}\text{Ar}/^{39}\text{Ar}$ ages older than corresponding muscovite $^{40}\text{Ar}/^{39}\text{Ar}$ ages, contrary to
15 the diffusion properties of these minerals, are common in the Himalaya and other metamorphic
16 regions. In these cases, biotite $^{40}\text{Ar}/^{39}\text{Ar}$ ages are commonly dismissed as ‘too old’ on account
17 of ‘excess Ar’. We present 32 step-heating $^{40}\text{Ar}/^{39}\text{Ar}$ ages from 17 samples from central
18 Himachal Pradesh Himalaya, India. In almost all cases, the biotite ages are older than predicted
19 from cooling histories. We document host rock lithology and chemical composition, mica
20 microstructures, biotite chemical composition, and chlorite and muscovite components of
21 biotite separates to demonstrate that these factors do not offer an explanation for the
22 anomalously old biotite $^{40}\text{Ar}/^{39}\text{Ar}$ ages. We discuss possible mechanisms that may account for
23 extraneous Ar (inherited or excess Ar) in these samples. The most likely cause for ‘too-old’
24 biotite is excess Ar, i.e. ^{40}Ar that is separated from its parent K. We suggest that this
25 contamination resulted from one or several of the following mechanisms: (1) ^{40}Ar was released

26 during Cenozoic prograde metamorphism; (2) ^{40}Ar transport was restricted due to temporarily
27 dry intergranular medium; (3) ^{40}Ar was released from melt into a hydrous fluid phase during
28 melt crystallization. Samples from the Main Central Thrust shear zone may be affected by a
29 different mechanism of excess-Ar accumulation, possibly linked to later-stage fluid circulation
30 within the shear zone and chloritization. Different Ar diffusivities and/or solubilities in biotite
31 and muscovite may explain why biotite is more commonly affected by excess Ar than
32 muscovite.

33

34 INTRODUCTION

35

36 In most applications, an $^{40}\text{Ar}/^{39}\text{Ar}$ mineral age is interpreted as the time since cooling
37 through its closure temperature (T_c) (Dodson, 1973; McDougall and Harrison, 1999). At
38 temperatures above T_c , Ar diffuses out of the mineral into an external reservoir; at
39 temperatures below T_c , Ar is quantitatively retained in the mineral. Dodson (1973) defined the
40 closure temperature as

$$41 \quad E/(R \times T_c) = \ln(A \times \tau \times D_0/a^2) \quad (1)$$

42 where E is the activation energy, D_0 is the diffusion coefficient, a is the radius of the effective
43 diffusion domain, A is the grain geometry factor, τ is the time constant, and R is the gas
44 constant. The concept of closure temperature is based on the assumptions that (1) the cooling
45 history is characterized by a linear increase in $1/T$; (2) Ar transport in the mineral is controlled
46 by volume diffusion; (3) Ar escapes into an ‘infinite’ reservoir, and the concentration of Ar at
47 the grain boundary remains zero (‘zero-concentration boundary condition’). Experimental
48 diffusion data suggest that the T_c of Ar in muscovite and biotite are $\sim 400\text{-}500$ °C and $\sim 300\text{-}$
49 400 °C, respectively, depending on grain size, mineral chemistry, and cooling rate (Harrison
50 et al., 1985; Grove, 1993; Grove and Harrison, 1996; Harrison et al., 2009).

51 Mineral $^{40}\text{Ar}/^{39}\text{Ar}$ ages may deviate from the inferred time of cooling through T_c for
52 several reasons. $^{40}\text{Ar}/^{39}\text{Ar}$ ages can be reset by neocrystallization or dynamic recrystallization
53 (e.g., Dunlap 1997; Mulch and Cosca, 2004). Hydrothermal fluids may displace radiogenic
54 ^{40}Ar through chemical reactions (e.g., Miller et al., 1991). Both mechanisms result in
55 $^{40}\text{Ar}/^{39}\text{Ar}$ ages that may be younger than the expected cooling ages, depending on the timing
56 of crystallization or fluid flow during cooling. In other cases, calculated $^{40}\text{Ar}/^{39}\text{Ar}$ ages are
57 ‘too old’; a short metamorphic pulse may be insufficient to completely degas Ar from a
58 mineral (e.g., Viete et al., 2011), or ^{40}Ar may become trapped, for example, in fluid inclusions
59 (Cumbest et al., 1994) or lattice defects (Camacho et al., 2012). The problem of anomalously
60 old $^{40}\text{Ar}/^{39}\text{Ar}$ ages is particularly known from biotite (e.g., Roddick et al., 1980; Baxter et al.,
61 2002) and commonly attributed to the presence of excess Ar.

62 ^{40}Ar from different sources may accumulate in a mineral. Slightly different
63 nomenclature is used throughout the literature; we follow the terminology of Dalrymple and
64 Lanphere (1969) and McDougall and Harrison (1999): Radiogenic Ar is ^{40}Ar produced within
65 the mineral by radioactive decay of ^{40}K . Inherited Ar is essentially radiogenic ^{40}Ar that
66 remained in the mineral during incomplete resetting (e.g., older core with younger rim) or was
67 introduced in the form of older material into the mineral (e.g., older K-bearing particles
68 become incorporated into younger volcanic rocks). Non-radiogenic Ar includes Ar of
69 atmospheric composition (here we use $^{40}\text{Ar}/^{36}\text{Ar} = 295.5$) and excess Ar. Excess Ar is parent-
70 less ^{40}Ar , i.e. ^{40}Ar that is separated from its K-bearing source; it is incorporated in rocks and
71 minerals by processes other than in-situ radioactive decay. Trapped Ar was incorporated
72 during mineral formation or at a later event and can encompass atmospheric Ar as well as
73 excess Ar. Inherited and excess Ar—collectively termed extraneous Ar—cause the age of the
74 mineral to appear older than its ‘true’ crystallization or cooling age.

75 In the Himalaya, many studies have reported biotite $^{40}\text{Ar}/^{39}\text{Ar}$ ages that were
76 considered ‘too old’ and were therefore excluded from the thermal history interpretations
77 (e.g., Hubbard and Harrison, 1989; Catlos et al., 2001; Stüwe and Foster, 2001; Godin et al.,
78 2006 and references therein; Horton et al., 2014; Adams et al., 2015). Other studies have
79 investigated possible sources of extraneous Ar and tried to link it, for example, to host-rock
80 composition (e.g., Foland, 1979; Boven et al., 2001; Baxter et al., 2002), presence or absence
81 of fluids (e.g., Cumbest et al., 1994; Stüwe and Foster, 2001; Itaya et al., 2009; Halama et al.,
82 2014), or (ultra)-high pressure metamorphism (e.g., Scaillet, 1998; Giorgis et al., 2000;
83 Warren et al., 2011). If extraneous Ar is quantifiable in the system, it may provide additional
84 information about the geologic history of the sample and the physicochemical conditions it
85 experienced (e.g., Kelley and Wartho, 2000).

86 With the goal of constraining the geological reason(s) behind anomalously old biotite
87 $^{40}\text{Ar}/^{39}\text{Ar}$ ages in the Himalaya and in metamorphic rocks in general, we present a dataset of
88 biotite and muscovite $^{40}\text{Ar}/^{39}\text{Ar}$ ages (BtAr, MsAr) from central Himachal Pradesh, NW
89 Himalaya. Petrographic microscope thin section observations, electron microprobe and X-ray
90 diffraction analyses, bulk-rock and mineral composition, and degree of biotite chloritization
91 were used to investigate structural or chemical relationships to the BtAr ages. Isochron
92 analyses of high-resolution step-heating experiments were applied to determine whether this
93 method could usefully detect the presence of excess Ar. The process we outline for
94 determining the likely sources of extraneous Ar are applicable to any metamorphic terrane.

95

96 **GEOLOGIC BACKGROUND**

97

98 In central Himachal Pradesh, Cenozoic sediments of the Himalayan foreland were
99 overthrust along the Main Boundary Thrust by the Proterozoic to early Cambrian Lesser

100 Himalayan Sequence (LHS). The Main Central Thrust (MCT), a middle to late Miocene
101 ductile shear zone, separates the LHS from the Greater Himalayan Sequence (GHS). The
102 GHS consists of lower greenschist to upper amphibolite facies Haimanta metagreywacke
103 (Neoproterozoic to Cambrian metapsammite and intercalations of metapelite and calcsilicate)
104 and Ordovician granite (Thöni, 1977; Steck, 2003; Webb, 2013; Figures 1 and 2). Dominant
105 foliation, metamorphic isograds and metamorphic zones based on index minerals (Bt + Chl,
106 Grt, Ky \pm St \pm Sil; Wyss, 2000; Steck, 2003; mineral abbreviations after Kretz, 1983) outline
107 a kilometer-scale, overturned to recumbent, SW-vergent antiform, resulting in an inverse
108 metamorphic sequence N and NE of the Kullu-Rampur window (overturned limb) and in a
109 normal metamorphic sequence in the Chandra valley (upper limb, northern study area) and S
110 and W of the town of Kullu (Figure 2; Thöni, 1977; Epard et al., 1995; Wyss et al., 1999).
111 The antiform is spectacularly exposed along the western hillslopes of the Beas valley ('Phojal
112 fold', Figure 2). Although the mechanism and tectonic setting are disputed, the folding is
113 generally attributed to early Miocene exhumation of mid-crustal rocks by SW-directed
114 thrusting and folding and accompanying erosion and/or normal faulting (e.g., nappe
115 emplacement, Epard et al., 1995; extrusion and channel flow, Searle et al., 2007; tectonic
116 wedging, Webb et al., 2007).

117 Along most of the Himalayan range, the hanging wall of the MCT is cut by the
118 normal-sense South Tibetan Detachment system (STD), which separates amphibolite-grade
119 metamorphic rocks of the GHS from the greenschist-facies to unmetamorphosed Tethyan
120 Himalayan Sequence. In the western Himalaya, the Zaskar Shear Zone and the Sangla
121 Detachment were identified as strands of the STD, but in central Himachal Pradesh, the
122 presence and location of shear zones equivalent to the STD are disputed (Thakur, 1998; Wyss
123 et al., 1999; Webb et al., 2007; Stübner et al., 2014; Figure 1). No strands of the STD system

124 are depicted in Figures 2A-C, although top-to-the-NE shear does occur, localized along the
125 contacts between intrusive and metamorphic rocks.

126 The highest-metamorphic grade rocks occur in the core of the Phojal fold (600-
127 700 °C, ~6 kbar; Epard et al., 1995; Leger et al., 2013), below the Deo Tibba Ordovician
128 granite (650-700 °C, ~8 kbar; Wyss, 2000), and in the deeply incised Chandra valley (Ky, St,
129 Hbl, Sil, indicating upper amphibolite facies; Epard et al., 1995; own observations). Here,
130 pegmatite and aplite dikes and veins suggest melting in the latest Eocene and Oligocene
131 (Wyss, 2000; Stübner et al., 2014). We will use the term ‘crystalline core’ to refer to these
132 amphibolite-grade metamorphic rocks (Ky ± Grt zones; Figures 2A-C). North and east of the
133 Kullu-Rampur window, the base of the Haimanta sequence above the MCT has experienced a
134 strong retrograde, greenschist-grade overprint with widespread chloritization of Bt and Grt
135 (Wyss, 2000; Figures 2A-C hatched area). Crustal thickening and prograde metamorphism
136 occurred in the Eocene and Oligocene (~41-26 Ma; Mnz and Grt geochronology; Walker et
137 al., 1999; Thöni et al., 2012; Stübner et al., 2014). The onset of exhumation is recorded by
138 decompression at 26-23 Ma, followed by rapid cooling to ~250 °C at ~30-60 °C/Ma, as
139 constrained by Ms and Bt Rb-Sr and ⁴⁰Ar/³⁹Ar and zircon fission-track (ZFT) ages (Schlup et
140 al., 2011 and references therein). Most MsAr ages are of 20-22 Ma (total of 20 samples from
141 Walker et al., 1999; Schlup et al., 2011; Stübner et al., 2014). Fewer BtAr dates are reported
142 in the literature, and are in general more variable and include contrasting ages (e.g., 42.6 ±
143 0.3 Ma and 16.7 ± 0.3 Ma from one locality with a MsAr age of 22.2 ± 0.2 Ma; Schlup et al.,
144 2011). Walker et al. (1999) and Webb et al. (2011) dismissed their BtAr results as
145 geologically meaningless on grounds of excess Ar.

146

147 **SAMPLES AND METHODS**

148

149 Seventeen samples from different lithologies and structural levels of the MCT hanging
150 wall in central Himachal Pradesh were collected for $^{40}\text{Ar}/^{39}\text{Ar}$ analysis (Table 1). Eleven Bt
151 and 14 Ms separates were analyzed at the Noble Gas Laboratory, Dalhousie University,
152 Halifax, Canada (DGC) (MsAr ages are published in Stübner et al., 2014). Additional three
153 MsAr and four BtAr ages were obtained using finely tuned step-heating experiments at the
154 Argon Lab, Freiberg, Germany (ALF) (Table 2). Analytical details of $^{40}\text{Ar}/^{39}\text{Ar}$ analysis at
155 both laboratories are given in the supplemental information.

156 Thin sections were investigated by optical microscopy to characterize the host rocks
157 and micas, with special attention to grain size, texture, and deformation textures such as
158 folding, kinking, undulose extinction, and fracture, indicating intra-crystalline strain
159 (Passchier and Trouw, 2005 and references therein). Backscatter electron (BSE) imaging at
160 high-contrast setting on JEOL JXA 8900 electron microprobes at the Universities of
161 Göttingen and Tübingen was used to characterize compositional variability of Bt in polished
162 thin sections. The same microprobes were used to determine chemical composition (Si, Ti, Fe,
163 Al, Na, K, Mg, Mn, Ca, Ba, Cr, F, Cl) of Bt by wavelength dispersive spectrometry (WDS),
164 using an acceleration voltage of 15 kV, a probe current of 15-20 nA, a beam diameter of 10-
165 20 μm , and counting times of 15 and 60 s on the X-ray signal and total background,
166 respectively. Both core and rim positions were analyzed. For separated grains, grain mounts
167 were used for WDS analysis with a probe current of 7 nA and a beam diameter of 1-2 μm .
168 Biotite chemical formulae were calculated stoichiometrically based on 22 oxygens. In order to
169 assess mineralogical purity of the Bt separates used for $^{40}\text{Ar}/^{39}\text{Ar}$ thermochronology, X-ray
170 powder-diffraction data were collected by the Rietveld method to quantify the amount of Chl
171 and Ms in Bt separates. Analytical procedures for Rietveld X-ray powder-diffractometry are
172 described in the supplemental information.

173

174 **RESULTS**

175

176 **Petrography**

177

178 Petrographic sample descriptions including photomicrographs are provided in the
179 supplemental information (Text and Figure S1). A brief summary is given below and in Table
180 1. Metasedimentary samples 807D1, 807E1, 811B1, 812D2, and 823G3 (Table 1) are medium
181 to coarse-grained micaschist to metapsammite with a typical assemblage of Qtz + Bt + Ms +
182 Pl ± Grt. Biotite is more abundant than Ms, aluminosilicates are not present, and accessory
183 minerals comprise Tur + Ap + Zrn ± Aln ± Mnz ± Ilm ± Rt ± Czo. Quartz is abundant in all
184 metasedimentary samples. Most samples show minor post-tectonic (across-foliation) growth
185 of Chl or chloritization of Bt and/or Grt. The foliation is pervasive, straight or wavy, and
186 parallel to compositional banding. Biotite lacks evidence of intracrystalline strain (e.g., kinks,
187 undulose extinction). Sample 807E1 has a mm-scale crenulation cleavage with Bt both along
188 the crenulation foliation and within microlithons; the Bt shows no evidence of intracrystalline
189 strain. Sample 814G1 is a fine-grained, banded metapsammite with abundant calcite; large
190 skeletal Ms grains at high angle to the foliation are probably detrital. Sample 827B1 is a fine-
191 grained metapelite with a wavy crenulation cleavage.

192 Granite and orthogneiss samples are quartz-rich and contain both Bt and Ms, except
193 for 823G2, which lacks Ms (Table 1). Garnet occurs in orthogneiss samples 823G2 and
194 819A2 and in leucogranite samples 803B3 and 815E1. Similar to the metasedimentary
195 samples, Bt shows no evidence of intracrystalline strain. In augengneiss samples 823G2 and
196 819A2, Bt forms the main foliation and is intergrown with Grt, suggesting no or little
197 deformation since (re)-crystallization of Bt + Grt. The same textures are observed in the
198 psammitic Haimanta metasedimentary rocks (e.g., 811B1).

199

200 **⁴⁰Ar/³⁹Ar Results**

201

202 Table 2 summarizes the ⁴⁰Ar/³⁹Ar results. Age spectra and inverse isochron plots are
203 shown in Figure 3, raw data are reported in Supplemental Tables S1 and S2. Many samples
204 yield high to medium percentages of atmospheric ⁴⁰Ar throughout the whole experiment
205 (Figure 3, Table S1). Therefore, the age spectra and the weighted mean ages (WMA)
206 calculated from all heating steps (e.g., Figure 3E, F) or the specified temperature range (e.g.,
207 Figure 3A, B) are strongly influenced by the atmospheric correction, such that even small
208 fluctuations in the initial ⁴⁰Ar/³⁶Ar ratio can cause relevant changes in the calculated step
209 ages. For most samples, the interpreted age (rounded values with ± 2σ error; Table 2) thus
210 relies on the inverse isochron age (IIA), which is unaffected by this type of correction.

211 A typical experiment at DGC comprised 12-16 heating steps. The age spectra of both
212 Ms and Bt often show complex patterns with only minor plateau sections (e.g., Figure 3B, K,
213 P, Q). Likewise, data in the inverse isochron plots are only partly arranged along single
214 mixing lines between trapped Ar (³⁶Ar/⁴⁰Ar intercept) and radiogenic Ar (³⁹Ar/⁴⁰Ar intercept;
215 e.g., Figure 3X, a, b, c). For two experiments, Ms of sample 814G1 and Bt of sample 827B1,
216 the scatter of data was too high to calculate reliable ages, and therefore probable age ranges
217 are suggested in the age interpretation (Figure 3V, g, Table 2). For other samples, IIAs were
218 calculated from all data points, or sometimes excluding the highest- or lowest-temperature
219 heating steps, and typically yielded mean square weighted deviates (MSWD) of ≤5 (Figure 3,
220 grey error boxes and ellipses and specified temperature range). Initial ⁴⁰Ar/³⁶Ar ratios range
221 from 210 to 420 and are mostly ≥295.5 (Table 2).

222 Additional Ms and Bt analyses, using a heating schedule of typically 18-20 steps, were
223 obtained at ALF. All analyses resulted in plateau ages that include 100% of the ³⁹Ar and with

224 MSWD <0.6 (Figure 3E, F, N, O, S, T, U; Table 2). These WMAs are identical to the IIAs,
225 which have MSWD <0.3. Initial $^{40}\text{Ar}/^{36}\text{Ar}$ ratios range from 270 to 340, and all lie within
226 uncertainty of the atmospheric value.

227 Muscovite ages of most samples range from 19 ± 1 to 23 ± 2 Ma (Table 2, Figures 2
228 and 3). Muscovite from sample 807E1—analyzed in both labs—yielded indistinguishable
229 ages (23 ± 2 and 22 ± 1 Ma; Figure 3L, N). Older Ms ages—up to 64 ± 5 Ma—originate from
230 lower-grade metamorphic rocks in the south and north of the study area (Figure 2A). Sample
231 807D1 yielded a complex age spectrum with step ages increasing from ~ 33 Ma to 102 Ma
232 with increasing temperature. An inverse isochron, fit to the lowest-temperature degassing
233 steps (800-900 °C), yielded 27.3 ± 0.7 Ma (Figure 3J). Muscovite ages in the Chandra valley
234 increase with elevation from 20 ± 1 Ma (807A1) through 24 ± 2 Ma (807C1) to 28 ± 5 Ma
235 (807D1; Figures 3E, G, J and 2A, elevation profile marked with white box frame).

236 The BtAr ages are generally older than the MsAr ages from the same locality
237 (compare e.g. Figures 3T and 3U). Most of them range from 20 ± 2 to 43 ± 2 Ma and are thus
238 up to 16 Ma older than the respective MsAr ages (Figures 2A and 4; Table 2). Samples from
239 the MCT zone (823G2, 823G3, and 827B1) yielded pre-Cenozoic BtAr ages (84 ± 6 Ma; 106
240 ± 5 Ma; Figure 2B). This inverted age relationship between Bt and Ms was observed
241 irrespective of lithology (c.f. Haimanta metasedimentary rocks vs. orthogneiss), structural
242 position (upper limb vs. core of the Phojal fold), and metamorphic zone (Ky vs. Grt zones;
243 Figures 2A, 2C and 4). The elevation profile in the Chandra valley records a positive age-
244 elevation trend (Figures 2A and 4). A ‘normal’ age relationship between Bt and Ms was
245 revealed for sample 804C1 from the Bt + Chl zone granite (Ms 64 ± 5 Ma, Bt 31 ± 2 Ma) and
246 for sample 807A1 from the Ky zone augengneiss (Ms 20 ± 1 Ma, Bt 20 ± 1 Ma).

247

248 **Biotite Chemistry**

249

250 Electron microprobe analyses of Bt revealed chemical compositions typical for
251 metamorphic rocks (Guidotti, 1984; Fleet et al., 2003), with 0.21–0.55 Mg/(Mg+Fe), 3.10–
252 3.47 Al per formula unit (p.f.u.), 0.15–0.33 Ti p.f.u., and low Mn, Ca, Na, Ba, and Cr (Table
253 3). F is <1.5 wt.%, Cl is close to or below the detection limit of 0.1 wt.%. Total oxides are 93-
254 97 wt.%. Within each sample, chemical compositions are narrowly defined without detectable
255 variations between Bt from different textural contexts, i.e., Bt within foliation, in Grt strain
256 shadows, from inclusions or embayments in Grt, with different grain size, or between cores
257 and rims. We recognized Chl and marginally chloritized Bt in BSE images in some of the
258 samples (see Table 1), but with no effects on bulk chemistry and with no correlation to the K-
259 content.

260

261 **Mineralogical Composition of Biotite Separates**

262

263 Powder XRD analyses suggest that the Bt separates used for $^{40}\text{Ar}/^{39}\text{Ar}$ dating comprise
264 84-98% Bt (Table 4, Figure 5). Trace amounts (0.6-2.4%) of Chl are present in all analyzed
265 samples; up to 15% Ms is intergrown with Bt in some samples. Repeated rinsing of the
266 separates in $\text{H}_2\text{O}_{\text{dest.}}$ reduced the Ms content (e.g., 812D2: 14% Ms in untreated separate vs.
267 5% Ms in rinsed separate; Figure 5), but a significant amount of Ms remained after rinsing
268 (e.g. 807E1: 15% Ms).

269

270 **Ti-in-Biotite Thermometry**

271

272 The Ti-in-biotite geothermometer of Henry et al. (2005) yielded Bt equilibration
273 temperatures of ~360-520 °C; 827B1 did not yield a valid result (not enough Ti, Table 3).

274 Regional trends in Ti-in-biotite temperatures correspond to the mapped metamorphic zones:
275 The highest temperatures (500-520 °C) were obtained from the center of the crystalline core
276 exposed in the Chandra valley (807E1, 807A1) and in the core of the Phojal fold (815C1);
277 lower temperatures (400-500 °C) were obtained from lower metamorphic-grade rocks (Grt ±
278 Ky zones); the lowest temperatures (<400 °C) were obtained from the structurally highest
279 samples in the upper limb of the Phojal fold (811B1, 807D1; Figure 2B). The elevation profile
280 in the Chandra valley records upsection decreasing temperatures (807A1, 807C1, 807D1;
281 Figure 2C).

282

283 **DISCUSSION**

284 **Do Muscovite and Biotite $^{40}\text{Ar}/^{39}\text{Ar}$ Ages Reflect Cooling through their Closure** 285 **Temperatures?**

286

287 In the metasedimentary rocks (807D1, 807E1, 811B1, 812D2, 815C1, 823G3)
288 foliation-forming Bt and Ms are interpreted to have grown during Oligocene prograde
289 metamorphism (see petrographic descriptions and Figure S1). Only the large skeletal Ms
290 grains in sample 814G1 are likely relict grains. In lower greenschist-grade metapelite 827B1,
291 some mica may be detrital, but most of the coarser grained, foliation-defining Bt and Ms are
292 interpreted as metamorphic. In the orthogneiss and augengneiss samples (806D3, 807A1,
293 807C1, 810B1, 819A1, 823G2), all mica appears to have recrystallized during the Cenozoic
294 tectonometamorphic cycle. In undeformed and weakly deformed granitic samples, mica grew
295 during magmatic crystallization, i.e., in the Ordovician (Bt and Ms in 804C1) or possibly the
296 Cenozoic (Ms in leucogranites 803B3 and 815E1). With the exception of the detrital grains,
297 there is no indication that one mica phase crystallized later than the other, e.g., as a result of

298 dynamic recrystallization in shear bands. The inverted age relationship between MsAr and
299 BtAr ages is therefore not a result of post-cooling neocrystallization or recrystallization of Ms.
300 Metamorphic and magmatic samples from the crystalline core, with 600-700 °C peak
301 metamorphic temperatures (Epard et al., 1995; Wyss, 2000), are expected to yield $^{40}\text{Ar}/^{39}\text{Ar}$
302 cooling ages (Warren et al, 2012). With increasing distance from the crystalline core, samples
303 are less likely to have resided at temperatures above Ar closure for a sufficiently long period
304 for complete thermal resetting. Figure 6 provides a compilation of all published as well as our
305 new thermochronologic ages from central Himachal Pradesh. Closure temperatures are $480 \pm$
306 $30 \text{ }^\circ\text{C}$ for MsAr and $350 \pm 30 \text{ }^\circ\text{C}$ for BtAr, calculated after equation (1) based on grain radii of
307 100-400 μm (Table 1), cooling rates of 20-100 $^\circ\text{C}/\text{Ma}$, and the diffusion parameters of
308 Harrison et al. (2009) and Grove and Harrison (1996). Monazite geochronologic data indicate
309 prograde metamorphism during ~41-26 Ma and onset of decompression between 26 and
310 22 Ma (Stübner et al., 2014 and references therein).

311 Rb-Sr and ZFT thermochronologic data from the crystalline core record early Miocene
312 cooling from 550 to 200 $^\circ\text{C}$ at an average rate of $\sim 20 \text{ }^\circ\text{C}/\text{Ma}$ (Figure 6, open red symbols and
313 arrow). The MsAr ages from the crystalline core (19-23 Ma) are consistent with these
314 independent thermochronologic and geochronologic constraints (Figure 6, red solid triangles);
315 they are younger than the Rb-Sr Ms ages and lie on the cooling path defined by the Rb-Sr and
316 ZFT chronometers. In contrast, most of the BtAr ages coincide with the timing of prograde
317 metamorphism and are 10-20 Ma older than what would be expected for cooling ages (Figure
318 6, red solid circles).

319 Samples from the northern (Chandra valley, Figure 6, blue symbols) and southern
320 study areas (black symbols) yield more variable Rb-Sr and ZFT ages and are, in general, a
321 few Ma older than those from the crystalline core. The MsAr ages range from ~ 20 to ~ 60 Ma
322 (Figure 6, black and blue solid triangles). The MsAr age spectrum of 807D1 (Figure 3G) may

323 represent a loss profile resulting from partial resetting at $\leq 28 \pm 5$ Ma (Forster and Lister,
324 2004; Viete et al., 2011). The MsAr age of 804C1 (64 ± 5 Ma; Figure 3A) predates the
325 Himalayan prograde metamorphism and may reflect partial resetting of Ms from this
326 Ordovician granite. The BtAr ages from these areas are—with two exceptions—5-10 Ma too
327 old to be interpreted as cooling ages (Figure 2).

328 Excluding the BtAr ages, the dataset indicates regional rapid exhumation and cooling
329 of the crystalline core since ~ 25 Ma (Figure 6). Age gradients between the younger crystalline
330 core and the slightly older lower-grade rocks may be attributed to diachronous exhumation.
331 The samples from the northern and southern area include rocks from the Grt and Bt + Chl
332 zones. There, residence at peak temperatures may have been insufficient (too short and/or too
333 cold) for complete resetting of the MsAr system, and these analyses may reflect partially reset
334 ages (e.g. 804C1, 803B3, 807D1, 827B1; Figure 6, blue and black solid triangles). Although
335 this interpretation implies that the BtAr ages from these samples are likewise partially reset, it
336 does not offer an explanation for the BtAr ages to be 5-20 Ma older than the corresponding
337 MsAr ages (Figure 2B). Therefore, we interpret the MsAr data as early Miocene cooling ages,
338 with the possible exception of a few samples from the low-grade metamorphic rocks (804C1,
339 803B3, 807D1, 827B1). BtAr ages are—with the possible exception of samples 814G1 and
340 807A1—not cooling ages.

341

342 **Do Biotite $^{40}\text{Ar}/^{39}\text{Ar}$ Ages Correlate with Biotite Equilibration Temperatures?**

343

344 The temperature and the portion of the pressure-temperature (P-T) path along which
345 Bt crystallized are important for determining links between the BtAr age and metamorphic
346 evolution. Although the Ti-in-biotite temperatures are consistent with regional metamorphic
347 zoning in the study area, the temperatures are generally 100-200 °C lower than the results

348 from Bt-Grt thermometry on samples from the same region (600-700 °C; Epard et al., 1995;
349 Wyss, 2000). Several samples do not meet the criteria for which the geothermometer was
350 calibrated, namely graphitic, peraluminous metapelites that contain ilmenite or rutile and have
351 equilibrated at ~4-6 kbar (Henry et al., 2005), suggesting that the temperature estimates may
352 be inaccurate. Biotite in metaluminous samples generally incorporates higher amounts of Ti
353 compared with peraluminous samples (Henry et al., 2005) therefore the calculated
354 temperatures may be upper estimates. Our Ti-in-biotite temperature estimates reveal two
355 characteristics (Figure 7A): (1) The oldest ArBt ages correspond to temperatures \leq ~400 °C.
356 (2) There is no correlation between ArBt age and Ti-in-biotite temperature above ~400 °C.

357

358 **Effects of Biotite and Host-rock Chemical Composition on Biotite $^{40}\text{Ar}/^{39}\text{Ar}$ Ages**

359

360 The Bt chemical composition of our samples is typical for high-Al metapelitic Bt
361 (Guidotti, 1984; Fleet et al., 2003) and similar to the samples used by Grove and Harrison
362 (1996) for the characterization of Ar diffusion in Bt (Table 3). In particular, Mg/(Fe+Mg)
363 ratios and anion compositions (F/(F+OH)) are comparable to our samples. Increased Ar
364 retentivity—proposed for F-rich and Fe-rich mica (Harrison et al., 1985; Grove and Harrison,
365 1996)—is thus an unlikely cause for the anomalously old Bt. Similarly, the other elements
366 analyzed (e.g., Ti, Cr, Mn, Ca, Ba, Na) do not show any unusual values. Although the Bt in
367 metasedimentary samples (807D1, 807E1, 812D2, 811B1, 814G1, 815C1, 823G3, 827B1)
368 have slightly higher Mg and lower F content compared to the magmatic samples (807A1,
369 807C1, 810B1, 823G2; Table 3), the differences are insignificant. Neither the samples that
370 yielded the oldest BtAr ages (823G2, 823G3, 827B1 \geq 100 Ma) nor those with BtAr ages
371 consistent with the regionally established cooling paths (807A1, 814G1) reveal any distinct
372 chemical characteristics.

373 Figure 4 shows BtAr and MsAr ages color-coded by rock type and sorted by their
374 location. There is no obvious correlation between the BtAr age and lithology. However,
375 chemical or mineralogical variations within each of the distinguished rock types are minor:
376 The magmatic rocks have 20-50% quartz, 15-40% plagioclase, 20-40% orthoclase;
377 leucogranite lacks biotite in contrast to granite/orthogneiss, which have biotite and muscovite
378 (see Kreidler, 2014 for chemical characterization of the magmatic rocks in central Himachal
379 Pradesh). The metasedimentary samples were obtained from the psammitic layers within the
380 greywacke sequence and are, therefore, quartz-rich, too. Thus, quartz as a potential local Ar-
381 sink (see discussion below) occurs in all sampled lithologies in similar proportions. Quartz-
382 poor metapelitic layers have not been sampled.

383 The discrepancy between the BtAr and MsAr ages tends to be larger in the southern
384 part of the study area compared to the north, suggesting that vicinity to the MCT shear zone
385 may be a factor responsible for the anomalous BtAr ages (Figure 4). This conclusion is further
386 supported by samples 823G2 and 823G3 from the MCT zone, which yielded Cretaceous BtAr
387 ages (despite Ti-in-biotite thermometry indicating Bt equilibration at ~430-490 °C during
388 Cenozoic metamorphism) and a MsAr cooling age from sample 823G3 at 22 ± 4 Ma (Table 2,
389 Figure 2A). However, distance to the MCT shear zone does not account for the anomalously
390 old Bt ages from the elevation profile in the Chandra valley, where BtAr ages increase faster
391 with elevation than the MsAr ages do (Figure 4).

392

393 **Effect of Chlorite on Biotite $^{40}\text{Ar}/^{39}\text{Ar}$ Ages**

394

395 The effect of chloritization on BtAr data is not well understood but it is known that
396 chloritization might severely disturb age spectra (e.g., Lo and Onstott, 1989). Handpicking of
397 Bt separates is an effective method to remove Chl and partly chloritized Bt in samples that

398 contain significant amount of Chl. However, our XRF data from four optically Chl-free Bt
399 samples show that ~2% of Chl remained in all separates even after multiple ultrasonic
400 cleaning of the grains (Figure 5, Table 4). Because there is no difference in Chl content
401 between the ‘well-behaved’ (807A1) and ‘too-old’ samples (807E1, 811B1, 812D2; Figure 5),
402 chloritization is unlikely the main cause for the anomalously old Bt ages. On the other hand,
403 samples 823G2, 823G3, and 827B1 from the MCT zone, which yielded pre-Cenozoic BtAr
404 ages, were strongly affected by retrograde metamorphism, accompanied by partial to
405 complete chloritization of Grt. We cannot exclude the possibility that chloritization of Bt
406 contributed to the anomalously old ages of these samples.

407

408 **Inherited Ar**

409

410 The Neoproterozoic to Cambrian Haimanta sedimentary rocks were intruded by
411 Ordovician (486 ± 6 Ma, Stübner et al., 2014) granite and affected by local contact
412 metamorphism and, possibly, a regional metamorphic overprint (Gehrels et al., 2003).
413 Minerals that have not been reset during Himalayan orogeny may therefore be as old as
414 ~500 Ma, and even small relicts of such old K-bearing minerals and fluid inclusions in
415 Himalayan metamorphic Bt may lead to too-old $^{40}\text{Ar}/^{39}\text{Ar}$ ages (Figure 8, point 2). The
416 probability that mineral phases and fluid inclusions with an early Paleozoic ^{40}Ar component
417 survive Cenozoic metamorphism is expected to decrease with the intensity of the
418 metamorphic overprint (e.g., Copeland et al., 1991; Viete et al., 2011; Mottram et al., 2015).
419 We argued earlier that in samples 804C1, 807D1, 814G1, and 827B1 the MsAr system and
420 therefore likely also the BtAr system may be partially reset, i.e. they may include a
421 component of inherited Ar. Older than expected BtAr ages, however, are observed throughout

422 the study area from the lowest to the highest-grade metamorphic zones (Figures 2B and 6),
423 suggesting that inherited Ar is unlikely to be the main cause of the too-old BtAr ages.

424

425 **Excess Ar**

426

427 After ruling out several factors that have previously been suggested to account for
428 anomalously old $^{40}\text{Ar}/^{39}\text{Ar}$ ages or reverse age relationships between MsAr and BtAr ages, we
429 suggest that excess Ar is the main factor producing too-old BtAr ages in the NW Himalaya.
430 Many previous $^{40}\text{Ar}/^{39}\text{Ar}$ studies of Himalayan rocks have come to a similar conclusion
431 sometimes leading authors to dismiss or not even present their BtAr ages (e.g., Hubbard and
432 Harrison, 1989; McFarlane, 1993; Vannay and Hodges, 1996; Walker et al., 1999; Godin et
433 al., 2001; Stüwe and Foster, 2001; Stephenson et al., 2001; Horton et al., 2014; Adams et al.,
434 2015), whereas other Himalayan studies report BtAr ages that are unaffected by excess Ar
435 (e.g., Hubbard and Harrison, 1989; Sorkhabi et al., 1993; Guillot et al., 1994; Wang et al.,
436 2006; Leloup et al., 2015).

437

438 ***Inverse Isochron Diagrams Do Not Reveal Excess Ar***

439

440 $^{40}\text{Ar}/^{39}\text{Ar}$ step-heating experiments offer, besides age determinations, the opportunity
441 to study the Ar degassing behavior of a specific mineral with increasing heating temperature.
442 In the simplest case, Ar isotopes in the mineral are a mixture between radiogenic and
443 atmospheric Ar, so that the data from the different heating steps plot in the inverse isochron
444 diagram along a straight mixing line with an intercept at the $^{36}\text{Ar}/^{40}\text{Ar}$ -axis defined by the
445 atmospheric Ar isotope composition (atmospheric $^{40}\text{Ar}/^{36}\text{Ar} = 295.5$). Excess Ar as a third
446 component becomes noticeable if it changes due to its non-atmospheric composition the

447 $^{36}\text{Ar}/^{40}\text{Ar}$ intercept (commonly stated as its reciprocal, the initial $^{40}\text{Ar}/^{36}\text{Ar}$ ratio). Usually,
448 excess Ar is enriched in ^{40}Ar , so that the initial $^{40}\text{Ar}/^{36}\text{Ar}$ ratio becomes higher. Increased
449 initial $^{40}\text{Ar}/^{36}\text{Ar}$ values are not uncommon in our experiments, for both Bt and Ms, with values
450 up to 553 ± 2 , but usually scattering between 300 and 350 (Table 2). No correlation between
451 inverse isochron ages and initial $^{40}\text{Ar}/^{36}\text{Ar}$ ratios is detectable, neither for all data together nor
452 for Ms or Bt alone (Figure 7B). Moreover, the high-resolution BtAr analyses from ALF, for
453 which all heating steps define a plateau age and which show significantly less scatter in the
454 inverse isochron diagrams (Figure 3E, F, N, O, S, T, U), yield trapped $^{40}\text{Ar}/^{36}\text{Ar}$ values
455 between 270 and 340, all indistinguishable within errors from the atmospheric value (Table
456 2).

457 The discussion about the occurrence of excess Ar and its detection from the Ar
458 degassing behavior during step-heating experiments is hampered by the fact that during step-
459 heating, excess ^{40}Ar may be released together with the radiogenic Ar, the trapped Ar or with
460 both (Kuiper, 2002). If only the trapped Ar component is contaminated with excess ^{40}Ar , the
461 $^{36}\text{Ar}/^{40}\text{Ar}$ intercept of the inverse isochron will yield initial $^{40}\text{Ar}/^{36}\text{Ar} > 295.5$, but the
462 $^{39}\text{Ar}/^{40}\text{Ar}$ intercept will reflect only the radiogenic ^{40}Ar , and the IIA will be unaffected by
463 excess Ar. Conversely, if only the radiogenic component is contaminated by excess ^{40}Ar , only
464 the age is changed, but not the initial $^{40}\text{Ar}/^{36}\text{Ar}$ ratio, even though the latter is usually regarded
465 as indication for excess Ar (Kuiper, 2002). This means that the non-existence of increased
466 initial $^{40}\text{Ar}/^{36}\text{Ar}$ ratios is not a proof for the absence of excess Ar. Therefore, excess Ar may
467 explain the too-old BtAr ages in the Himachal Pradesh despite that fact that initial $^{40}\text{Ar}/^{36}\text{Ar}$
468 ratios are not consistently higher than the atmospheric ratio.

469

470 ***Which Processes May Have Lead to Excess Ar in the Biotite?***

471

472 The $^{40}\text{Ar}/^{39}\text{Ar}$ age dating technique relies on the assumption that Ar concentrations
473 outside of the mineral grain remain close to zero, so that Ar that diffuses out of the mineral
474 grain escapes into an ‘infinite reservoir’ (Figure 8, point 1; e.g., Dodson, 1979). This reservoir
475 may be a local sink, such as a fluid or a host rock mineral with a high Ar solubility, or an
476 external reservoir—ultimately the atmosphere. The ‘total local sink capacity’ is a function of
477 the modes of mineral and fluid phases in the vicinity of the source mineral and their respective
478 Ar partition coefficients; specifically, it has been suggested that the presence or absence of
479 quartz in a rock affects the total local sink capacity and hence controls excess Ar
480 accumulation (Baxter, 2003). In addition, an efficient Ar transport from the Ar-releasing
481 mineral grain into the external reservoir is a critical aspect in $^{40}\text{Ar}/^{39}\text{Ar}$ thermochronology
482 (e.g., Baxter, 2003). Transport of Ar in a rock volume depends on the presence or absence of
483 fluids in the intergranular space, and it is necessary to distinguish ‘open systems’, in which
484 fluid circulation facilitates rapid transport of Ar over large distances (meters to kilometers),
485 and dry ‘closed systems’, in which transport of Ar is limited to a few centimeters over
486 millions of years (Scaillet, 1996; Kelley, 2002; Baxter, 2003). In dry systems, ^{40}Ar may
487 accumulate along grain boundaries and partition into minerals resulting in ‘internally’ derived
488 excess Ar; in fluid-rich systems, if the fluid is enriched in ^{40}Ar , partitioning of ^{40}Ar into the
489 mineral may result in ‘externally’ derived excess Ar (see Kelley, 2002 and references therein;
490 Baxter, 2007). Here we consider the following (not mutually exclusive) options for excess Ar
491 accumulation:

- 492 1. High ^{40}Ar partial pressure in the intergranular medium (Figure 8, point 3).
- 493 2. Dry intergranular medium and/or Cenozoic partial melting disturbing the ^{40}Ar balance
494 (Figure 8, points 4 and 5).
- 495 3. Fluid enriched in ^{40}Ar resulting in excess Ar incorporation into biotite (Figure 8, point 3).

496

497 ***High ⁴⁰Ar partial pressure in the intergranular medium***

498 Biotite and other K-bearing minerals from the Neoproterozoic/Cambrian Haimanta
499 metagreywacke and/or Ordovician granite broke down and/or outgassed during Eocene-
500 Oligocene prograde Barrovian metamorphism, releasing radiogenic ⁴⁰Ar (⁴⁰Ar*) that had
501 accumulated over ~500 Ma. A typical Haimanta rock with 3-4 wt% K₂O (Vance and Mahar,
502 1998; Chambers et al., 2009; Stübner, unpublished data) could result in ~100 ppb ⁴⁰Ar* in the
503 host rock, assuming ⁴⁰Ar* accumulation over 500 Ma and no Ar loss (see Appendix 1). For
504 the granite with 6 wt% K₂O (Kreidler, 2014) the ⁴⁰Ar* concentrations would add up to ~200
505 ppb in a closed system. These ⁴⁰Ar* concentrations are 10-20 times higher than the ~11 ppb
506 ⁴⁰Ar* that would have accumulated in a Bt with 10 wt.% K₂O over ~20 Ma, and could
507 significantly disturb the ⁴⁰Ar* budget of Bt and other mineral phases. High Ar partial pressure
508 in the intergranular space in combination with the lower Ar solubility in Ms than Bt may
509 result in partitioning of ⁴⁰Ar into Bt in preference to Ms (Figure 8, point 3) (see Kelley, 2002
510 for a recent summary of Ar solubility). This mechanism predicts that metasedimentary and
511 magmatic rocks are both affected by excess Ar, depending on their respective mineralogical
512 composition (e.g., quartz content) and K concentration (Baxter, 2003). This may explain why
513 both magmatic and metasedimentary samples are affected by excess Ar (Figure 4), but it does
514 not provide a straightforward explanation why BtAr ages from the MCT zone are significantly
515 more affected by excess Ar than samples from the MCT hanging wall.

516

517 ***Dry intergranular medium and Cenozoic partial melting disturb the ⁴⁰Ar balance***

518 Solubility of Ar in hydrous fluids is 4-5 orders of magnitude higher than in most
519 minerals (see compilation in Kelley, 2002) providing an effective local sink for Ar as well as
520 serving as pathway to a more external sink (Baxter, 2003; Baxter et al., 2007; Figure 8, point
521 1). For dry systems, it has been shown both experimentally (Baxter et al., 2007) and in field

522 studies (Foland, 1979; Scaillet, 1996) that restricted mobility of Ar ensues accumulation of
523 ⁴⁰Ar in the intergranular medium and can result in excess Ar in minerals.

524 In the Himachal Himalaya, there is ample evidence for fluid circulation throughout the
525 Cenozoic. Examples are: (1) Ubiquitous quartz segregation veins are associated with early to
526 late Himalayan deformation and metamorphism (e.g., Epard et al., 1995; Wyss et al., 1999).
527 (2) Tourmaline is one of the main accessory phases in the Haimanta metasedimentary rocks.
528 (3) Ky + Qtz segregation veins are common and indicate the presence of aqueous fluids
529 during amphibolite-facies metamorphism. (4) Pegmatite dikes intruded along the axial
530 surfaces of folds are related to the late stages of thrusting along the MCT and formation of the
531 Phojal fold in the late Oligocene-early Miocene (Epard et al., 1995; own observations). We
532 therefore consider it unlikely that dry conditions existed throughout the entire Cenozoic
533 metamorphic cycle.

534 It is, however, conceivable that partial melting and melt extraction consumed hydrous
535 fluids, resulting in locally dry pore spaces (Figure 8, point 4). The NW Himalaya was affected
536 by a protracted history of crustal melting between ~36 and 18 Ma (e.g., Dèzes et al., 1999;
537 Robyr et al., 2006; Langille et al., 2012; Lederer et al., 2013; Stübner et al., 2014), similar to
538 other locations in the Himalaya (e.g., Rubatto et al., 2013; Zeiger et al., 2015). Removal of
539 hydrous fluids by late Eocene-Oligocene migmatization could have significantly reduced the
540 total local sink capacity (Baxter, 2003), resulting in accumulation of excess Ar. However,
541 most if not all of our Bt samples are from rocks that did not undergo partial melting; for dry
542 conditions to have affected the BtAr ages, migmatization and melt extraction must have
543 affected the fluid budget on a several-kilometer scale. In this case, it is theoretically possible
544 that the excess-Ar Bt ages provide a maximum age estimate of migmatization.

545 Silicate melts have Ar solubilities intermediate between those of hydrous fluids and
546 most minerals (e.g., Kelley, 2002 and references therein). During migmatization, melts are

547 likely to serve as an additional sink for intergranular ^{40}Ar . During melt crystallization, ^{40}Ar
548 may be released back into the intergranular medium, increasing its ^{40}Ar concentration.
549 Consequently, excess Ar may diffuse from the intergranular medium into the mineral phases
550 for as long as the rocks reside at temperatures above T_c (Figure 8, point 5). Some Himalayan
551 Miocene leucogranite yields geologically reasonable BtAr ages, reflecting the timing of
552 emplacement and cooling (e.g., Copeland et al., 1990; Hodges et al., 1998; Wang et al., 2006),
553 whereas in other locations, the BtAr ages are apparently affected by excess Ar (Horton et al.,
554 2014). Therefore, migmatization does not offer a straightforward explanation for excess BtAr
555 ages in the study area or in the Himalayan orogen in general.

556 Migmatization and melt emplacement may be related to accumulation of excess Ar in
557 minerals through two effects: (1) It affects the availability of hydrous fluids in the pore space,
558 and (2) the high solubility of Ar in melt affects the distribution of ^{40}Ar in melt, hydrous fluid
559 and mineral phases. A better understanding of these mechanisms requires determination of
560 partition coefficients of Ar between silicate melt, hydrous fluids, and mineral phases, in
561 particular for Ms and Bt.

562

563 ***Fluid circulation along the MCT***

564 In the NW Himalaya, the MCT was active between ~26-23 Ma and ~17-15 Ma (e.g.,
565 Stephenson et al., 2001; Vannay et al., 2004). This thrust activity was associated with intense
566 fluid circulation, causing retrograde greenschist-facies metamorphism with chloritization of
567 Bt and Grt and formation of cm- to dm-sized quartz veins within the ~3 km wide shear zone
568 (e.g., Wyss, 2000). Circulation of fluids has been documented along the entire MCT shear
569 zone, for example, in Nepal (Copeland et al., 1991; Evans et al., 2008; Derry et al., 2009) and
570 Bhutan (Stüwe and Foster, 2001; Mottram et al., 2015), where it was proposed to account for
571 excess Ar in Bt and a reverse relationship between MsAr and BtAr ages. In central Himachal

572 Pradesh, two MsAr ages from the MCT shear zone and its immediate hanging wall are
573 geologically reasonable: 823G3 (22 ± 4 Ma) is indistinguishable from other MsAr cooling
574 ages from the hanging wall crystalline rocks; 827B1 (32 ± 9 Ma) is consistent with other
575 thermochronometric ages from the same locality (Figure 2A). In contrast, Bt from these
576 samples yielded pre-Cenozoic $^{40}\text{Ar}/^{39}\text{Ar}$ ages, significantly older than any other BtAr ages
577 from the study area and predating Cenozoic prograde metamorphism.

578 Studies on the composition of modern hydrothermal fluids along the MCT suggest a
579 large component of meteoric water (e.g., Derry et al., 2009), but the Ar isotopic composition
580 of these fluids is unknown. High partial pressures of Ar of any isotopic composition can slow
581 or inhibit diffusive loss of radiogenic ^{40}Ar from the mineral, thus leading to accumulation of
582 internally derived excess Ar (e.g., Baxter, 2003). In the end-member scenario of high Ar
583 partial pressure in the intergranular medium blocking diffusional escape of radiogenic ^{40}Ar ,
584 the excess Ar Bt age could theoretically record the time when the intergranular medium
585 became saturated in Ar. Therefore, the pre-Cenozoic BtAr ages from three samples from the
586 MCT zone (827B1, 823G2, 823G3) cannot be attributed to an influx of atmospheric Ar in the
587 late Oligocene-Miocene alone. If anomalous BtAr ages from the MCT zone result from fluid
588 circulation within the MCT shear zone, as suggested by the spatial correlation in the study
589 area and by $^{40}\text{Ar}/^{39}\text{Ar}$ thermochronology studies in other localities in the Himalaya (Hubbard
590 and Harrison, 1989; Stüwe and Foster, 2001), these hydrous fluids must have been enriched in
591 ^{40}Ar (Figure 8, point 3). The source of this ^{40}Ar may, for example, be prograde metamorphic
592 Ar release (see above). Possible mechanisms leading to excess Ar in Bt include partitioning of
593 ^{40}Ar from the fluid phase into the mineral or trapping in fluid inclusions. Because the Bt
594 samples from the MCT zone are an order of magnitude more strongly affected by excess Ar
595 than the hanging-wall samples (Cretaceous vs. Cenozoic BtAr ages), and excess Ar in the
596 hanging wall samples does not vary systematically with distance to the MCT (Figure 4), we

597 suggest that various, although possibly related mechanisms account for excess Ar in the MCT
598 zone and in the hanging wall, respectively.

599

600 **CONCLUSIONS**

601

602 Most of the BtAr ages yielded from granitic and metapelitic samples from Himachal
603 Pradesh are older than MsAr ages from the same samples and 5-20 Ma older than the
604 expected timing of cooling through BtAr closure temperature derived from independent
605 geochronologic evidence. Apart from BtAr ages from the MCT zone, which are pre-Cenozoic,
606 BtAr ages tend to cluster at 20-40 Ma. The 'too-old' BtAr ages do not correlate with lithology
607 or structural position. The Bt separates contain only ~2 % Chl, suggesting that chloritization is
608 not the main cause for these anomalously old BtAr ages.

609 We suggest that the likely cause for the 'too-old' BtAr ages in the Himachal Pradesh is
610 excess Ar in Bt but not, or to a lesser extent, in Ms. Possible sources of excess Ar are:

611 (1) ^{40}Ar released from older K-bearing minerals during Cenozoic prograde metamorphism and
612 mineral breakdown, which partitioned into Bt and/or inhibited diffusive loss of radiogenic
613 ^{40}Ar at high temperature;

614 (2) ^{40}Ar that accumulated in Bt at temperatures above T_c because a temporarily dry
615 intergranular medium, possibly resulting from late Eocene-Oligocene migmatization and melt
616 extraction, restricted ^{40}Ar transport towards an external sink;

617 (3) ^{40}Ar released from melt into a hydrous fluid phase during Oligocene melt crystallization
618 resulting in high ^{40}Ar concentrations in the rock and partitioning of ^{40}Ar into Bt.

619 Samples from the MCT shear zone yield significantly older BtAr ages than samples
620 from the hanging wall rocks and may be affected by a different mechanism of excess-Ar

621 accumulation, possibly linked to fluid circulation within the MCT zone and strong
622 chloritization.

623 The proposed mechanisms of excess-Ar accumulation in Bt depend on the solubility
624 of Ar in minerals, hydrous fluids and melts, and further experiments that determine partition
625 coefficients between these phases will provide further insight into the problem of excess Ar;
626 in particular, although it is accepted that solubility of Ar in minerals is an order of magnitude
627 lower than in hydrous fluids, the partition coefficients between different mineral phases is
628 poorly known (see compilation in Kelley, 2002). Excess Ar is more commonly detected in Bt
629 than in Ms, possibly due to higher solubility of Ar in Bt compared to Ms. This makes Bt more
630 susceptible to excess-Ar accumulation if the zero-Ar boundary condition is violated in an
631 open or closed system. The higher susceptibility of Bt to excess Ar compared to Ms could
632 also be due to its diffusion characteristics, with its higher Ar diffusivity responsible for
633 incorporation of more excess Ar in cases where a surplus is available in the intergranular
634 medium. This surplus can be provided by Ar-enriched fluids or might be an effect of Ar
635 accumulation under dry conditions (closed system), but it can also be caused by an increasing
636 partition coefficient between mineral and fluid during post-metamorphic cooling (Kelley,
637 2002). Decreasing solubility of Ar in fluids at decreasing temperatures triggers its diffusion
638 into the solid phase, preferentially into minerals with high Ar diffusivity and thus more
639 readily into Bt compared to Ms. These considerations suggest that in most sample sets, in
640 which Bt is affected by excess Ar it is most likely that Ms, too, contains excess Ar, although
641 its concentrations are probably smaller and may not affect $^{40}\text{Ar}/^{39}\text{Ar}$ ages.

642

643 **APPENDIX 1. Calculation of ^{40}Ar accumulation**

644 Assuming 3.5 wt% K_2O in Haimanta sediments and using molar masses $\text{K}_2 = 2 \times 39.1$
645 g/mol and $\text{K}_2\text{O} = 94.2$ g/mol, the amount of K in Haimanta sediments is $3.5 \times (2 \times 39.1 /$

646 94.2) = 2.91 wt% K₂. Using the molar fraction of ⁴⁰K/K = 0.000117 the amount of ⁴⁰K in the
647 sediment is 0.000117 × (2.91/100) × 1E+6 = 3.4 ppm ⁴⁰K or 3.4 mg ⁴⁰K per 1 kg rock. The
648 decay of ⁴⁰K to ⁴⁰Ar* is calculated after McDougall and Harrison (1999, Eq. 2.11). For t =
649 500 Ma the amount of ⁴⁰Ar* is 0.114 ppm (0.114 mg ⁴⁰Ar per 1 kg rock). For Ordovician
650 granite with typically 6 wt% K₂O (Kreidler, 2014), the same calculation yields 0.684 ppm
651 ⁴⁰Ar accumulated over 500 Ma.

652

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664

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924

925 **FIGURE CAPTIONS**

926 Figure 1. Overview tectonic map of the NW Himalaya (modified from Stübner et al., 2014).

927 Black rectangle corners outline the extent of Figure 2.

928

929 Figure 2. (A) Geologic map of the study area with own and published thermochronology ages

930 (in $\text{Ma} \pm 2\sigma$ error). Isograds and metamorphic zones are after Steck (2003) and own

931 observations. In the western part of the study area, published isograds are redrawn to reflect

932 the geometry of the Phojal fold, which folds the metamorphic zones (Epard et al., 1995).

933 $^{40}\text{Ar}/^{39}\text{Ar}$ ages are from this study (Table 2); Rb-Sr ages from Mehta (1977); zircon fission

934 track (ZFT) from Schlup et al. (2011; superscript S) and Walia et al. (2008; superscript W).

935 White outer frame denotes samples from elevation profile near Rohtang pass. (B) Simplified

936 version of Figure 2A showing sample numbers and biotite $^{40}\text{Ar}/^{39}\text{Ar}$ ages (in $\text{Ma} \pm 2\sigma$). (C)

937 Biotite $^{40}\text{Ar}/^{39}\text{Ar}$ ages ($\text{Ma} \pm 2\sigma$) projected onto a SSW-NNE profile with 2-times vertical

938 exaggeration illustrating the large-scale, SW-vergent, recumbent fold in the hanging wall of
939 the Main Central Thrust (MCT). The fold axis of the MCT dips to the NW, therefore the
940 structural position of samples NW and SE of the profile are not accurately reflected by their
941 true elevation above sea level. For samples 827B1, 814G1, 823G2, 823G3, and 819A1, the
942 approximate structural position relative to the MCT and the recumbent fold rather than the
943 sample elevation is shown.

944

945 Figure 3. Age spectra and inverse isochrons of $^{40}\text{Ar}/^{39}\text{Ar}$ analyses; bold labels in lower left
946 indicate sample number and mineral (Ms, muscovite; Bt, biotite), labels in lower right indicate
947 laboratory where analysis was conducted (DGC, Noble Gas Laboratory, Dalhousie, Halifax,
948 Canada; ALF, Argon Lab, Freiberg, Germany). Muscovite $^{40}\text{Ar}/^{39}\text{Ar}$ data analysed at DGC
949 are from Stübner et al. (2014). Weighted mean ages (WMA) and inverse isochron ages (IIA)
950 are calculated from all data points and/or from subsets of consecutive heating steps that fit a
951 single regression line (grey error boxes and grey error ellipses); for each sample, the same sets
952 of heating steps were used for WMA and IIA calculation. Note that for some samples (e.g.,
953 807D1 Ms, 810B1 Ms, 827B1 Ms) the initial $^{40}\text{Ar}/^{36}\text{Ar}$ is significantly different from
954 atmospheric composition (295.5), and therefore the IIA, which is independent from
955 atmospheric correction, is the more reliable age compared to the WMA, which is calculated
956 assuming atmospheric initial $^{40}\text{Ar}/^{36}\text{Ar}$. MSWD is the mean square of weighted deviates;
957 prob. is the probability of fit (for detailed definitions and discussion, see Ludwig, 2008).

958

959 Figure 4. Structural position (top) and lithology of the samples together with their biotite and
960 muscovite $^{40}\text{Ar}/^{39}\text{Ar}$ ages. Background shading and letters indicate generalized lithology.
961 Small box outlines six ages from an elevation profile near Rohtang pass (marked by outer
962 white box on Figure 2A, B). Biotite $^{40}\text{Ar}/^{39}\text{Ar}$ ages of samples 823G2 and 823G3 are 106 ± 5

963 Ma and 84 ± 6 Ma, respectively, and therefore plot outside the diagram. ALF, ArgonLab
964 Freiberg; MCT, Main Central Thrust.

965

966 Figure 5. Mineralogical composition of biotite separates, determined by Rietveld X-ray
967 diffractometry, together with their $^{40}\text{Ar}/^{39}\text{Ar}$ ages ($\pm 2\sigma$). Separates were obtained by magnetic
968 separation and handpicking to remove non-biotite phases; ‘rinsed’ samples were further
969 purified by repeated rinsing in deionized H_2O in an ultrasonic bath. All samples dated at ALF
970 were rinsed. Note that y-axis starts at 80%, not at zero.

971

972 Figure 6. Thermochronologic data (own and published) and regional cooling path for the
973 study area (see Figure 2 for geographic locations and references). Yellow and hatched grey
974 bands indicate closure temperatures with respective uncertainties. Red arrow indicates the
975 approximate cooling path of the crystalline core (red symbols).

976

977 Figure 7. (A) Ti-in-biotite temperatures vs. biotite $^{40}\text{Ar}/^{39}\text{Ar}$ ages ($\pm 2\sigma$). (B) Initial $^{40}\text{Ar}/^{36}\text{Ar}$
978 ratios vs. inverse isochron ages (both $\pm 2\sigma$) as listed in Table 2.

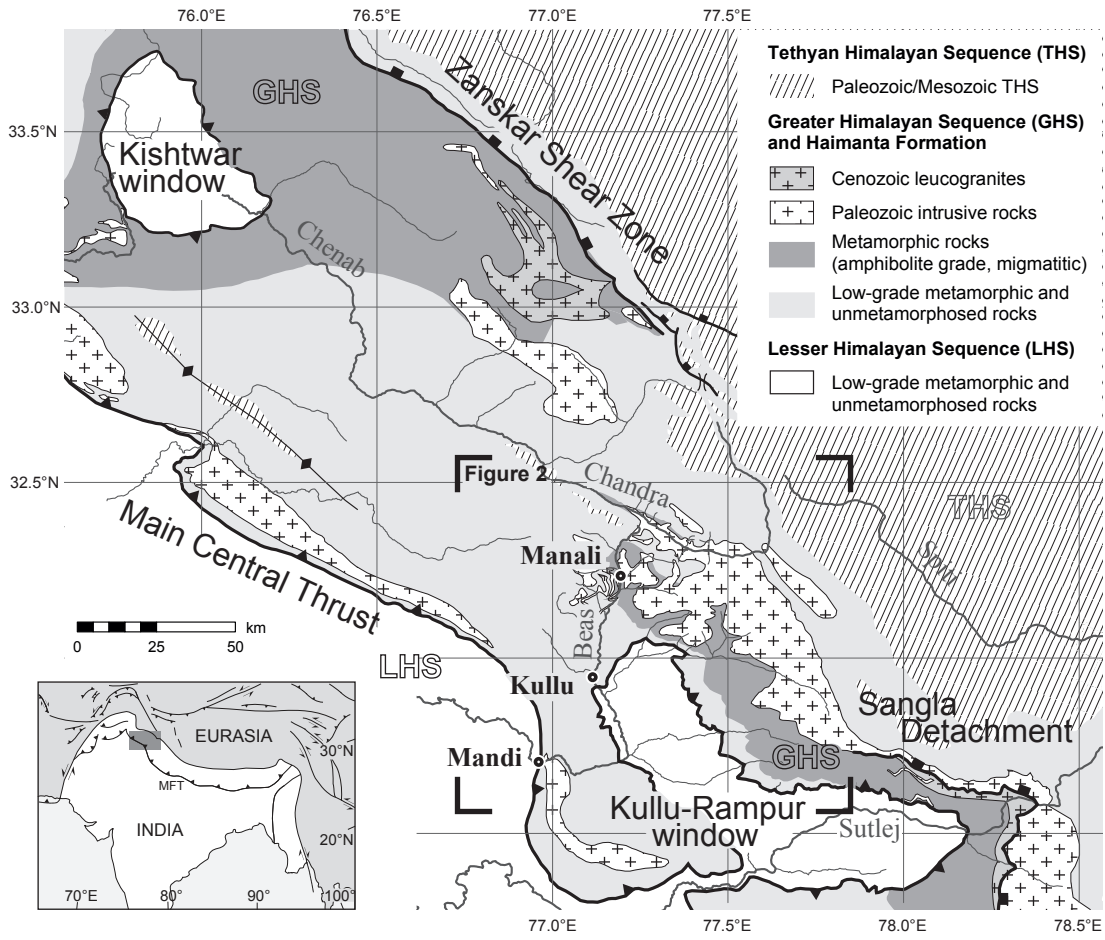
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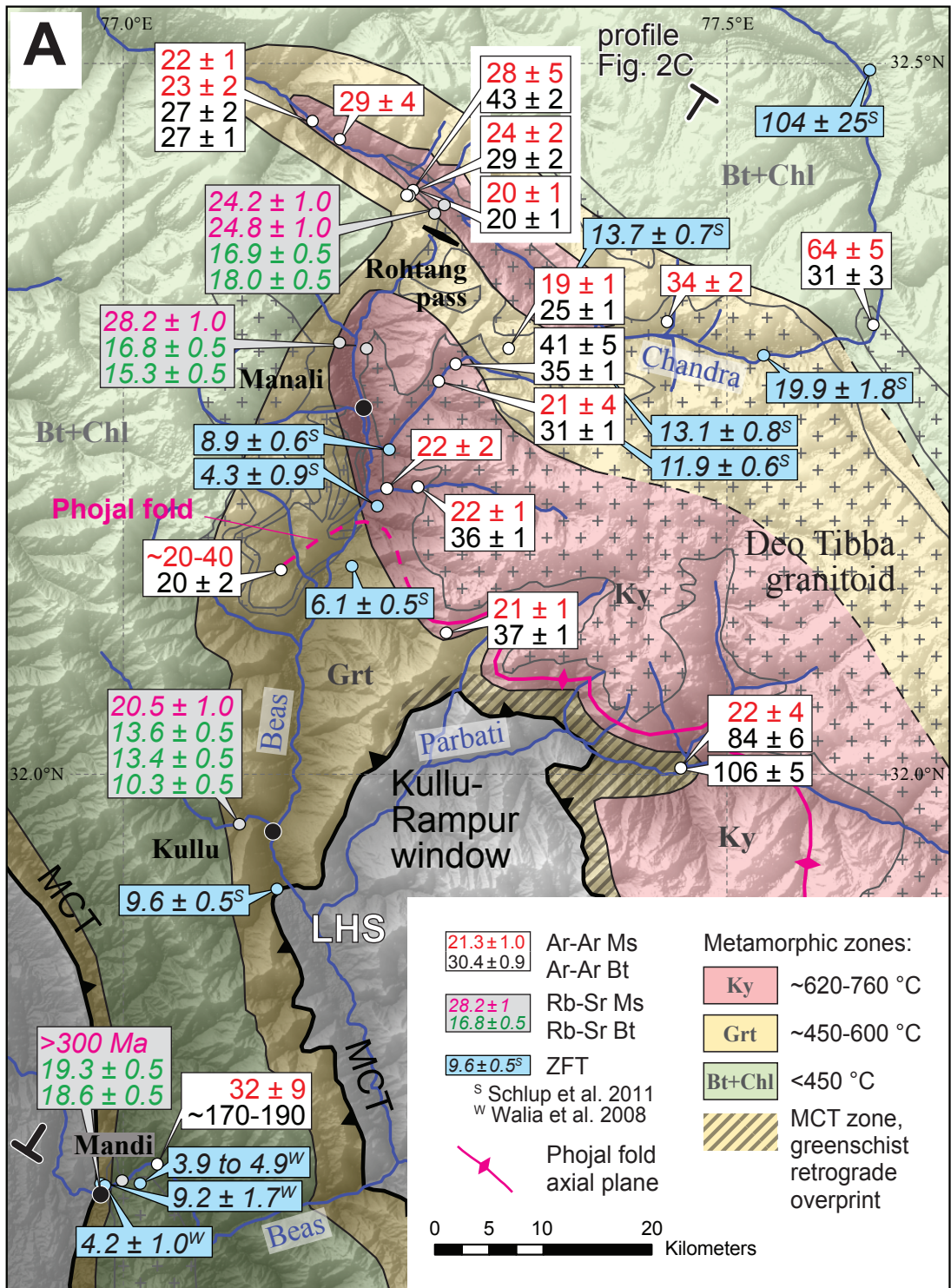
980 Figure 8. Schematic diagram illustrating Ar production and movement in and around a biotite
981 grain above its closure temperature. The biotite grew or recrystallized dynamically during a
982 metamorphic cycle. See text for details.

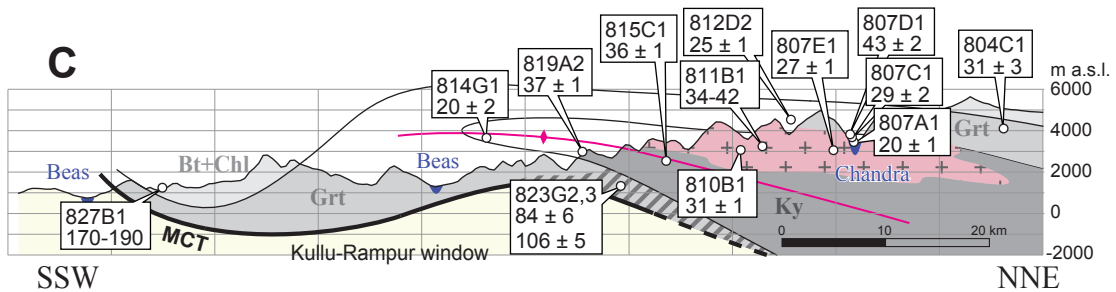
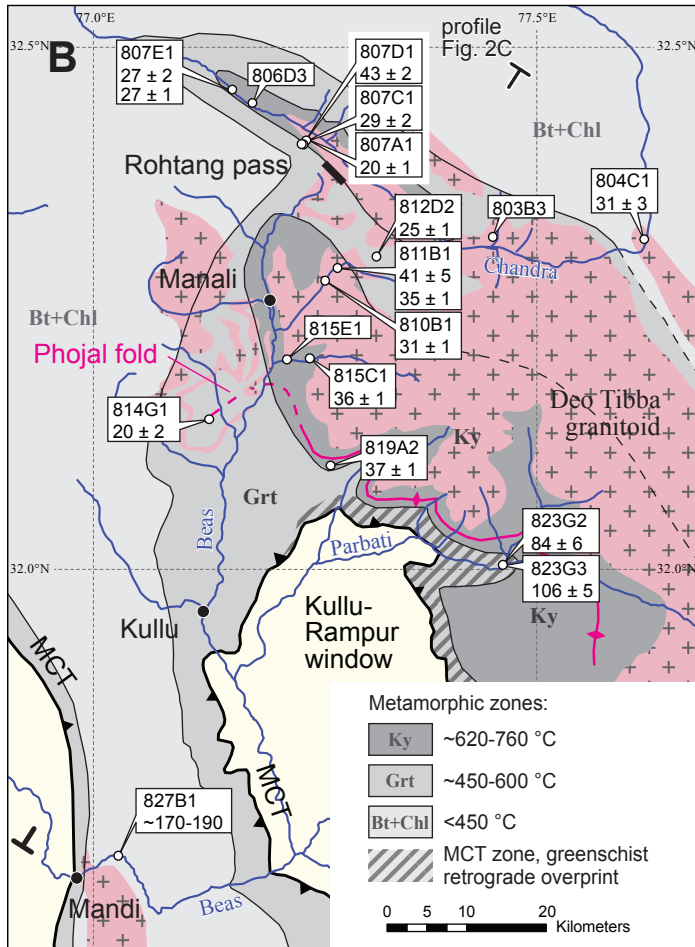
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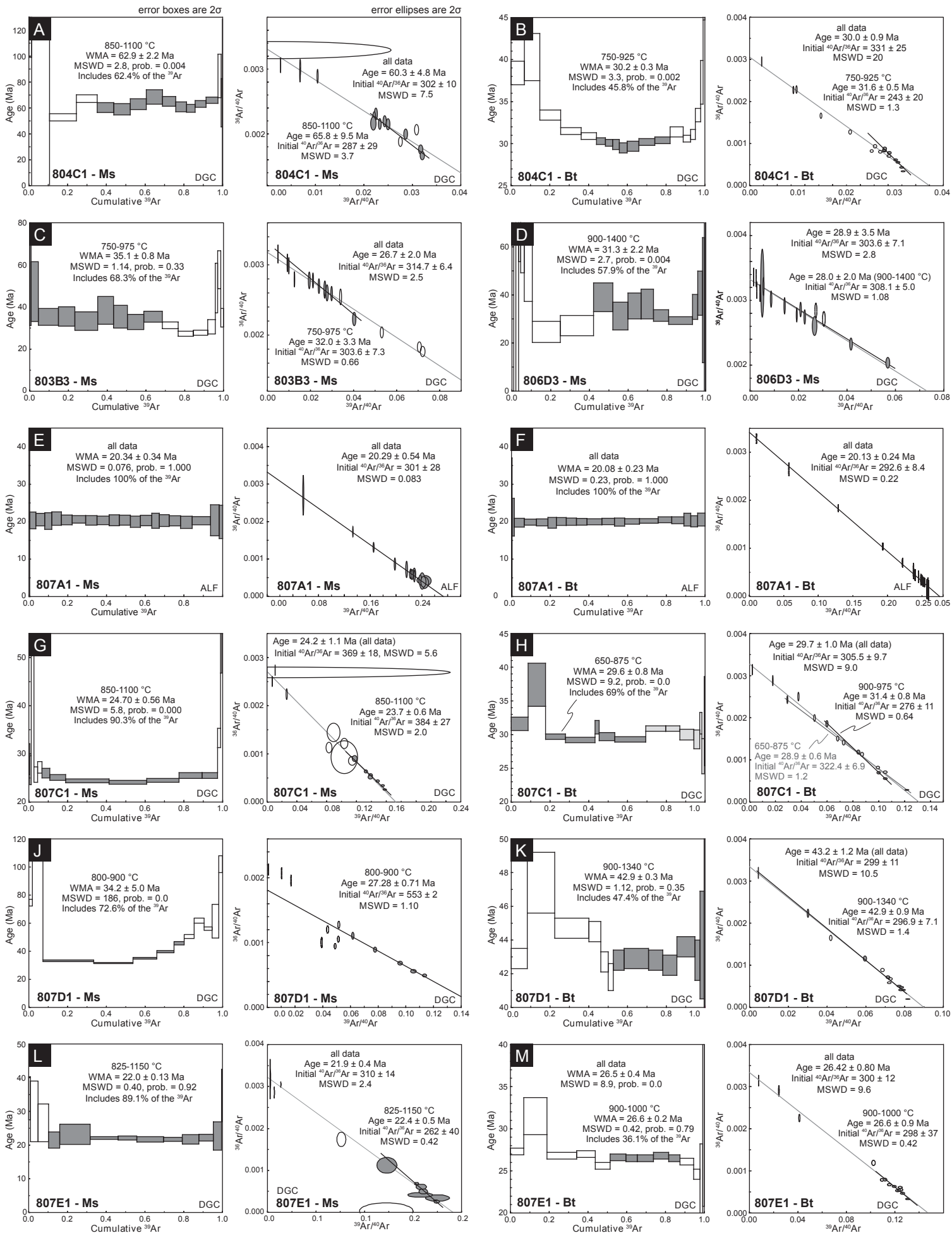
984 ¹GSA Data Repository item 201Xxxx, including $^{40}\text{Ar}/^{39}\text{Ar}$ raw data (Tables S1, S2), thin
985 section descriptions and photomicrographs (supplemental text and Figure S1), and analytical
986 details of $^{40}\text{Ar}/^{39}\text{Ar}$ analyses and Rietveld X-ray powder-diffractometry (supplemental text) is
987 available online at www.geosociety.org/pubs/ft20XX.htm, or on request from

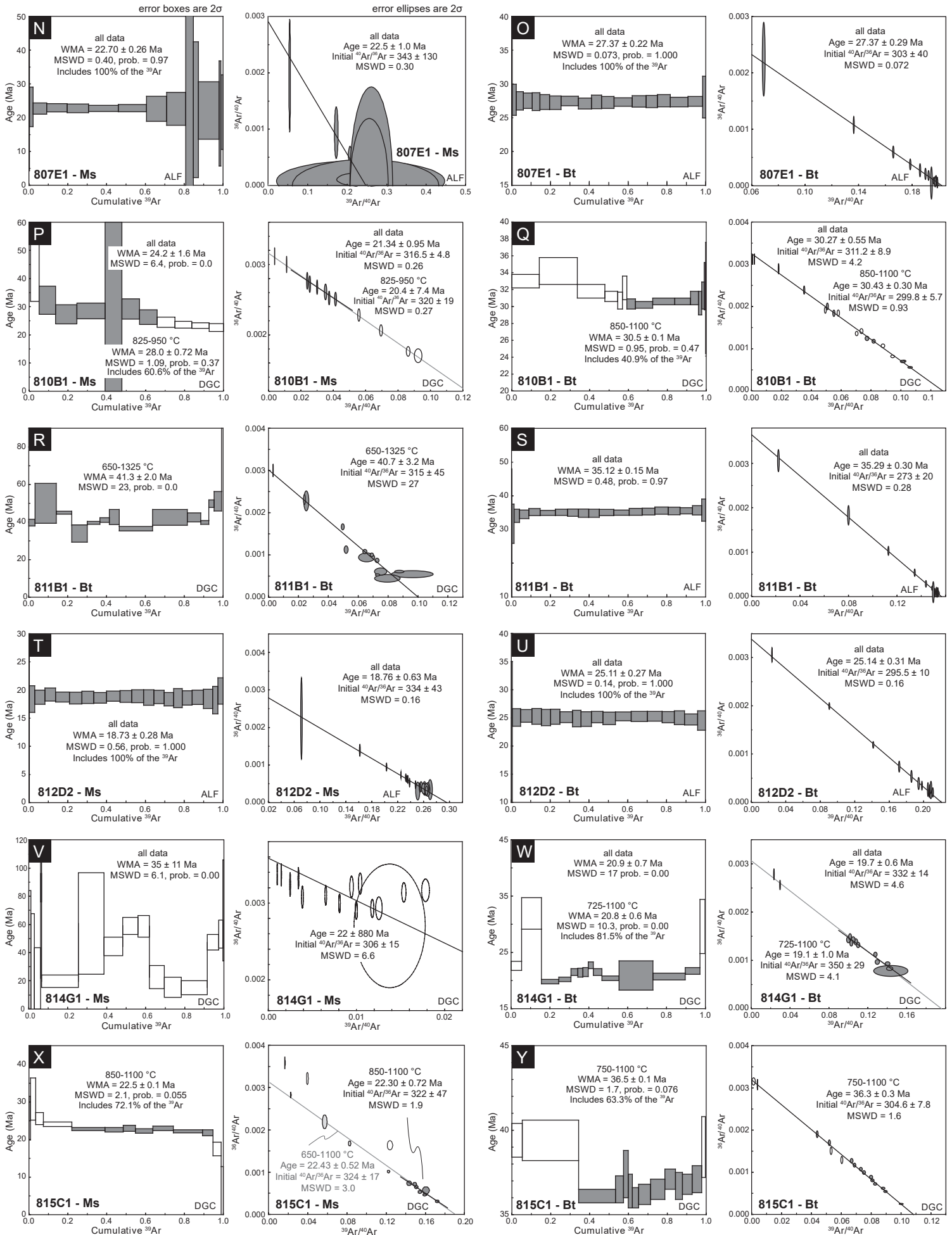
988 editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301,
989 USA.

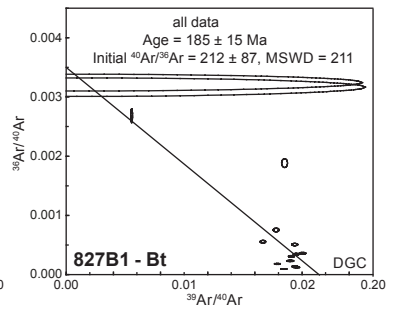
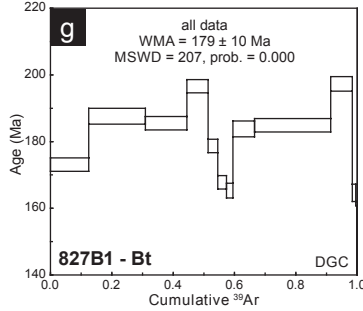
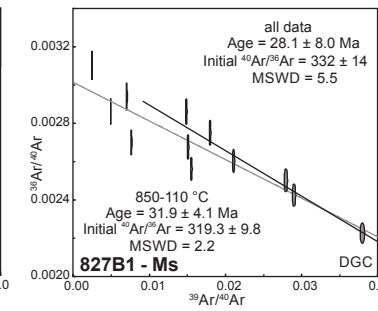
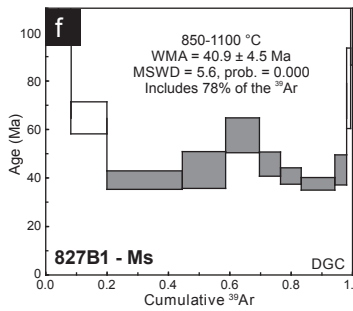
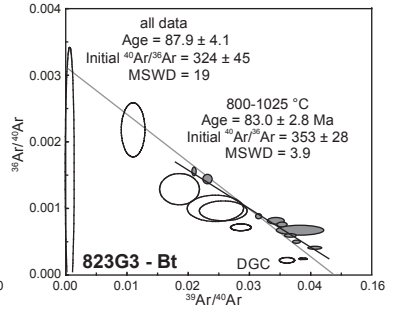
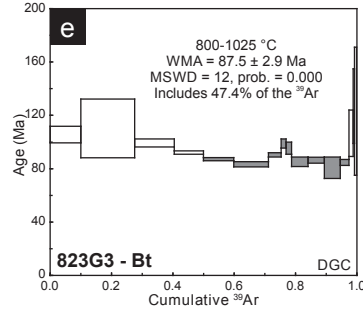
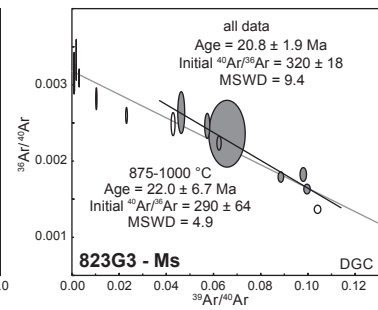
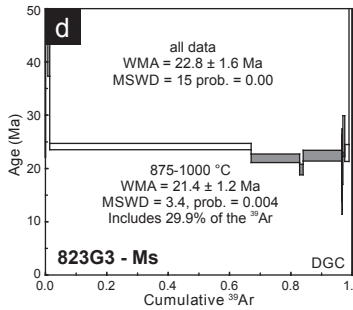
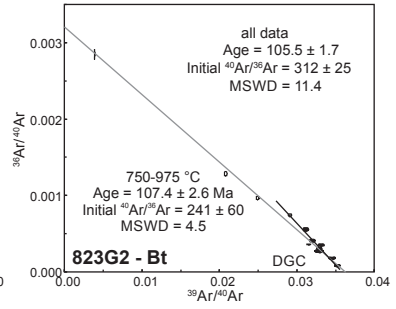
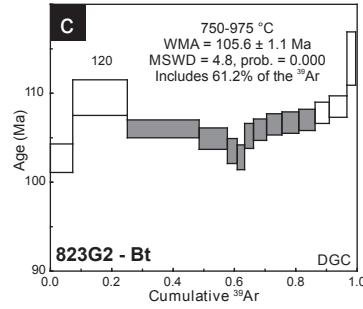
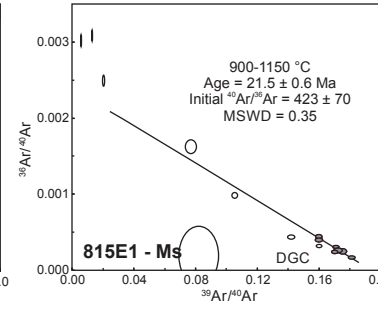
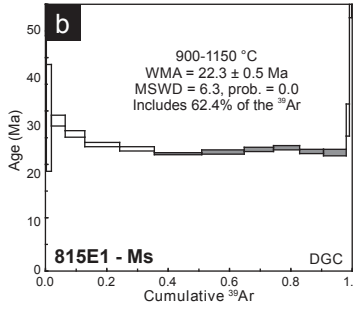
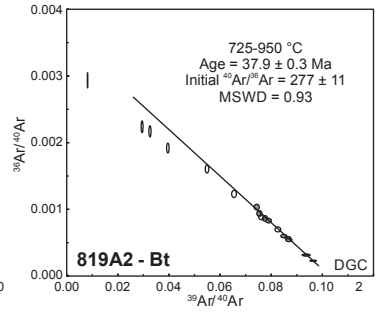
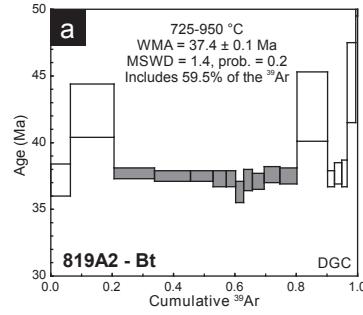
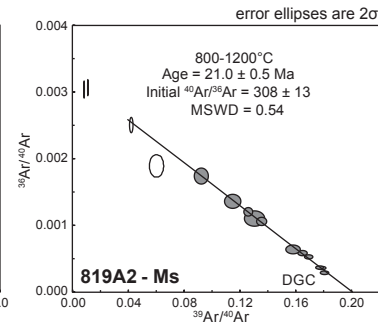
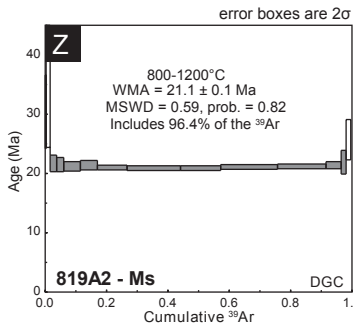


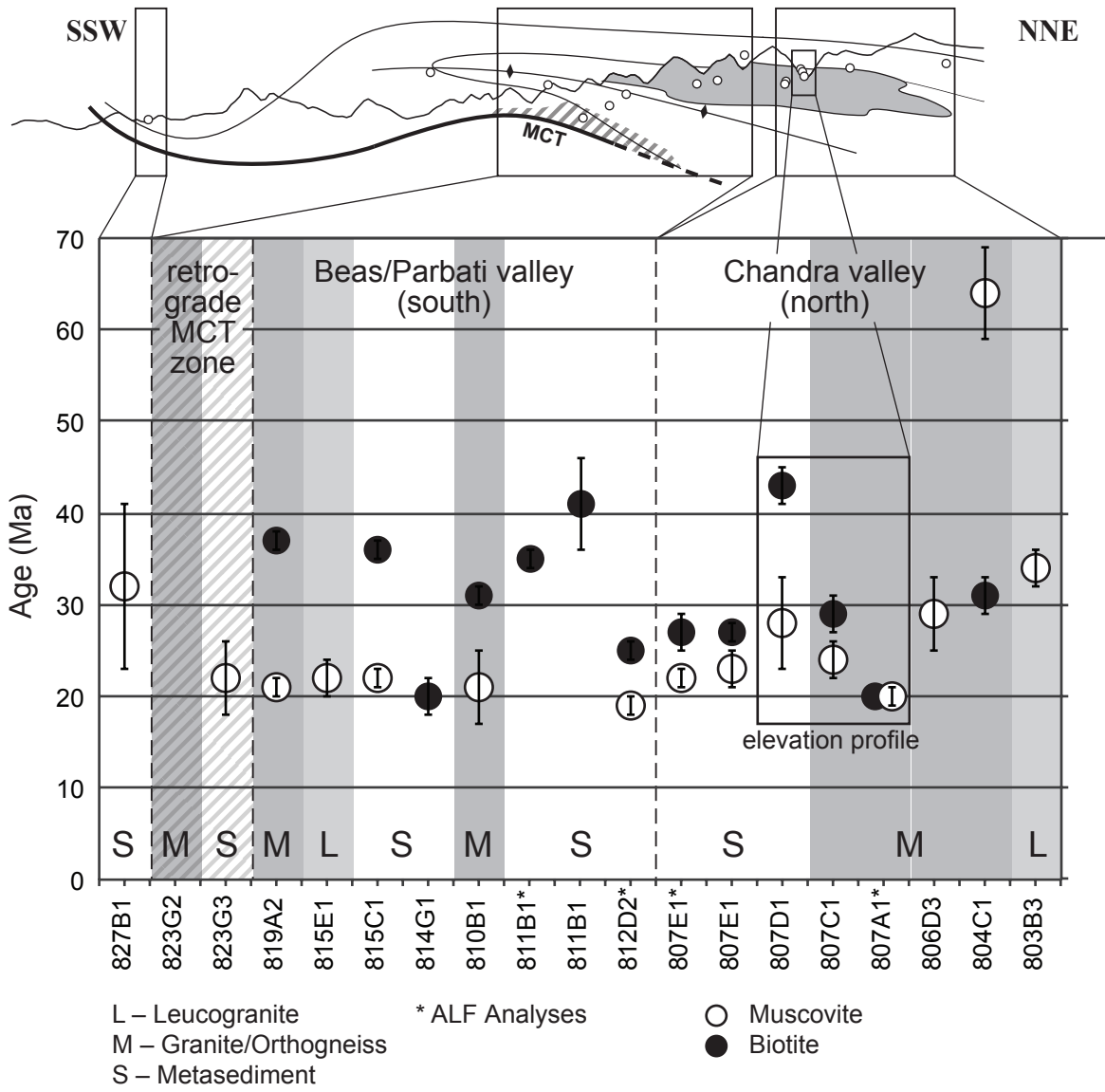


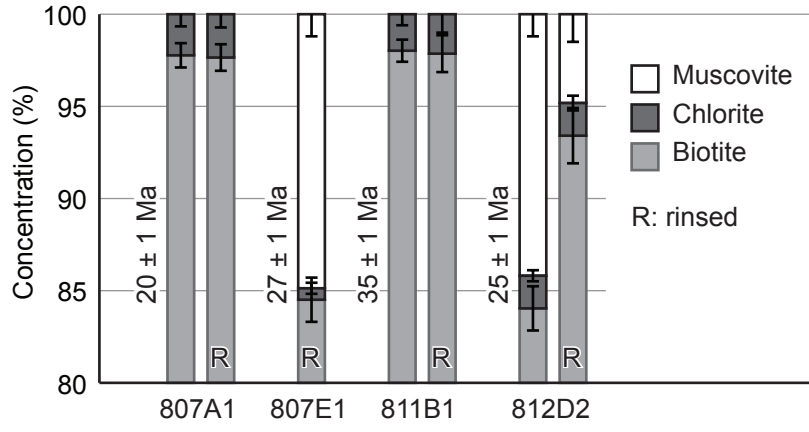


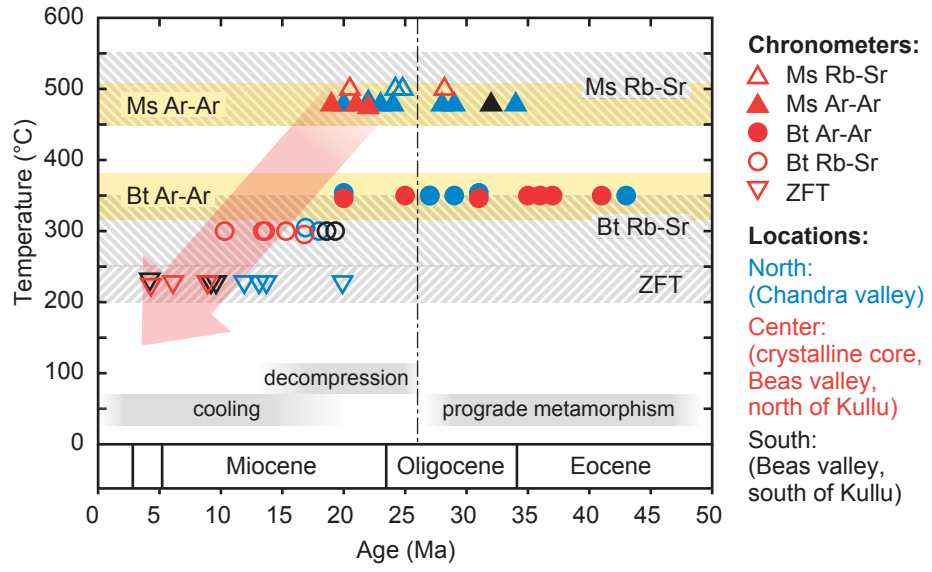


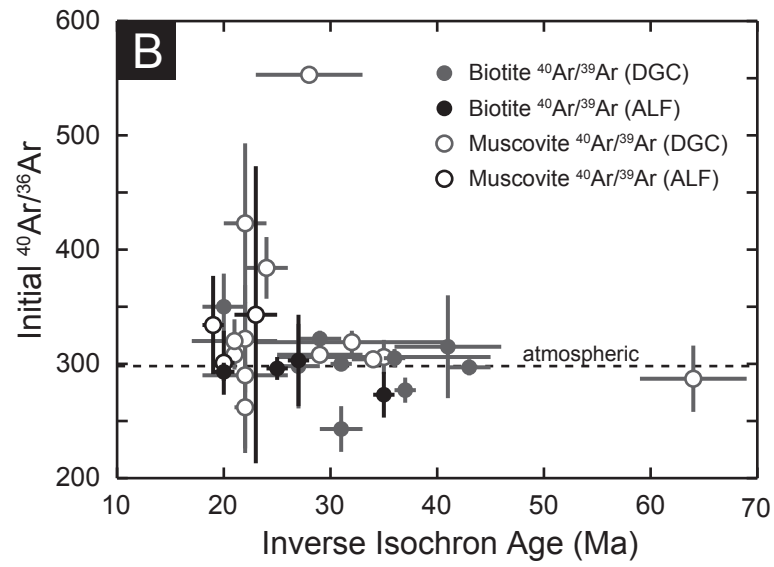
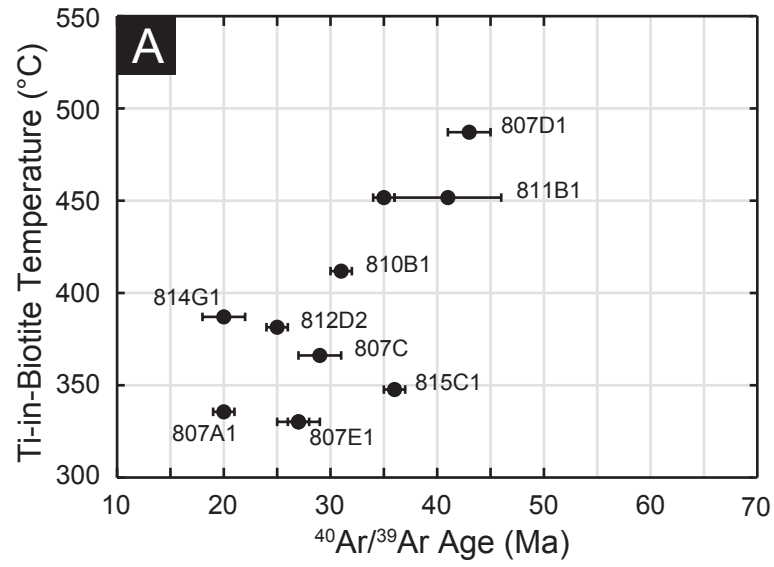












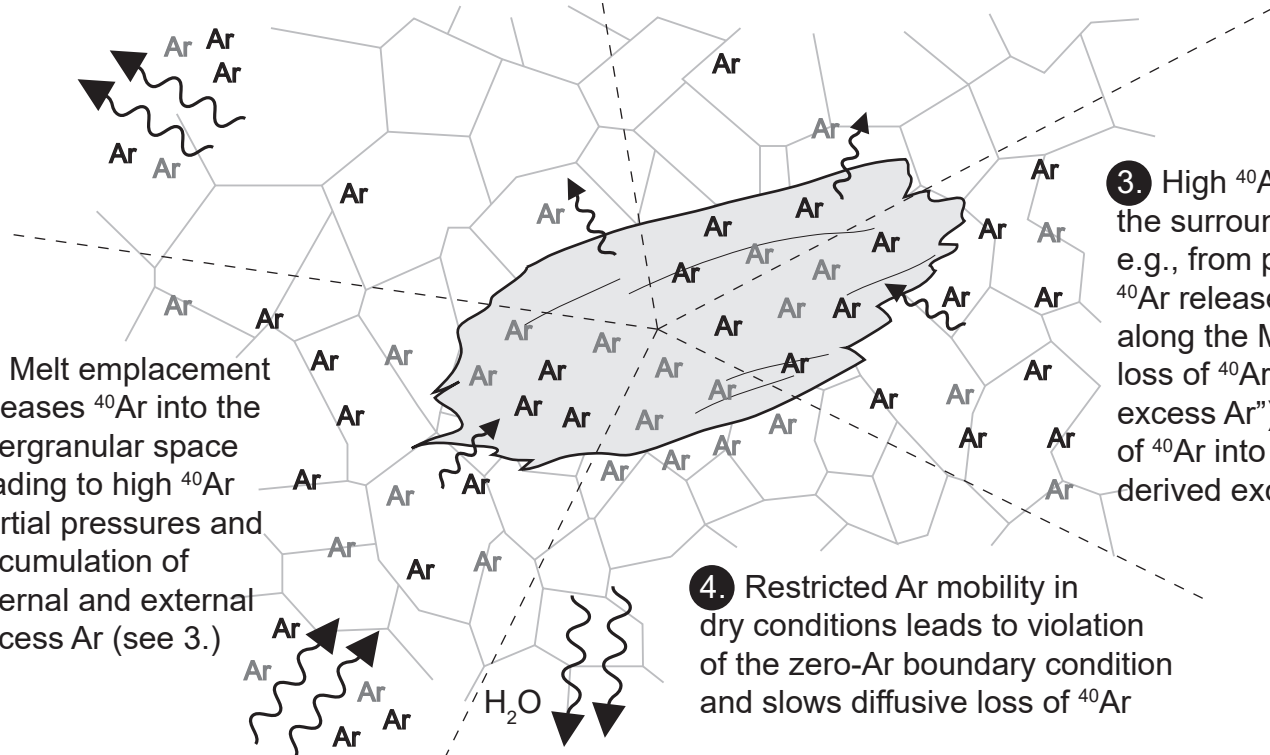
1. ^{40}Ar released during prograde metamorphism escapes from the system: no accumulation of excess Ar

2. Inherited ^{40}Ar is trapped, for example, in mineral inclusions or retained in the lattice of the incompletely degassed pre-Cenozoic biotite ("partially reset age")

5. Melt emplacement releases ^{40}Ar into the intergranular space leading to high ^{40}Ar partial pressures and accumulation of internal and external excess Ar (see 3.)

3. High ^{40}Ar partial pressure in the surrounding rock volume, e.g., from prograde metamorphic ^{40}Ar release or from fluid circulation along the MCT, slows diffusive loss of ^{40}Ar ("internally derived excess Ar") and facilitates diffusion of ^{40}Ar into the grain ("externally derived excess Ar")

4. Restricted Ar mobility in dry conditions leads to violation of the zero-Ar boundary condition and slows diffusive loss of ^{40}Ar



Migmatization and melt extraction lead to consumption of hydrous fluids and (temporarily) dry conditions

Ar ^{40}Ar produced before Cenozoic biotite growth or recrystallization

Ar ^{40}Ar produced in biotite grain since its Cenozoic (re-)crystallization

Ar may be present in dry or fluid-filled intergranular space and in biotite and other K-bearing and K-free mineral phases according to the respective Ar solubilities

Wiggly arrows indicate diffusion of ^{40}Ar into and out of the biotite grain (small arrows) and fluid (Ar, H₂O) exchange between the intergranular space surrounding the biotite and an external reservoir (large arrows)