1 **Geochronology of accessory allanite and monazite in the Barrovian**

2 **metamorphic sequence of the Central Alps, Switzerland**

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16 **Abstract**

17   The formation of accessory allanite, monazite and rutile in amphibolite facies-

18 rocks across the Barrovian sequence of the Central Alps (Switzerland) was investigated

19 with a combination of petrography and geochemistry and related to the known

20 structural and metamorphic evolution of the Lepontine dome. For each of these minerals

21 a specific approach was adopted for geochronology, taking into account internal zoning

22 and U-Th-Pb systematics. In-situ U-Th-Pb dating of allanite and monazite by ion

23 microprobe revealed systematic trends for the ages of main deformation and

24 temperature in the Lepontine dome. Isotope dilution TIMS dating of rutile returns dates

25 in line with this picture, but is complicated by inheritance of pre-Alpine rutile and

26 possible Pb loss during Alpine metamorphism.

27   Allanite is generally a prograde mineral that is aligned along the main foliation of

28 the samples and found also as inclusions in garnet. Prograde allanite formation is

29 further documented by rutile inclusions with formation temperatures significantly

30 lower than the maximum T recorded by the rock mineral assemblage. Allanite ages vary

31 from 31.3 ± 1.1 Ma in orthogneisses in the East to 31.7 ± 1.1 Ma for a Bündnerschiefer

32 and 28.5 ± 1.3 Ma for a metaquartzite in the central area, to 26.8 ± 1.1 Ma in the western

33 part of the Lepontine dome. These ages are interpreted to date the main deformation

34 events (nappe stacking and isoclinal deformation of the nappe stack), close to peak

35 pressure conditions.

36   The timing of the thermal peak in the Lepontine dome is recorded in monazite that

37 grew at the expense of allanite and after a main episode of garnet growth at

38 temperatures of ~620°C. Monazite in the central area yields an age of 22.0 ± 0.3 Ma,

39 which is indistinguishable from the age of 21.7 ± 0.4 Ma from a metapelite in the

40   western part of the Lepontine dome. In the central area some of the classical kyanite-

41   staurolite-garnet schists directly underlying the metamorphosed Mesozoic sediments

42   contain monazite that records only a pre-Alpine, Variscan metamorphic event of upper

43   greenschist to lower amphibolite facies-conditions dated at ~330 Ma.

44   The new age data provide evidence that nappe stacking at prograde amphibolite-

45   facies conditions and refolding of the nappe stack occurred between 32 and 27 Ma, only

46   a few million years after eclogite-facies metamorphism in the Adula-Cima Lunga unit.

47   Amphibolite-facies metamorphism lasted for about 10 My to ~22 Ma, allowing for

48   multiple ductile deformation and recrystallization events. The long lasting amphibolite-

49   facies metamorphism requires fast cooling between 20 and 15 Ma in the Central Alps.

50   This fast cooling was not related to an increase in sedimentation rates in the foreland

51   basins, suggesting that tectonic exhumation was responsible for termination of

52   amphibolite-facies metamorphism in the Lepontine dome.

53

1. Introduction

Well-constrained geochronology is essential for understanding metamorphic processes that occur during orogenesis. Not only are ages of metamorphic events important for establishing and correlating the timing of orogen-scale processes but equally important is the rate at which these processes occur. Finding evidence of a prograde history, crucial for evaluating models of heat transfer during orogenesis, is often difficult because such evidence within a sample is usually obliterated by subsequent, higher-grade metamorphism. Therefore, geochronology of Barrovian metamorphic terranes is best achieved by combining different geochronometers that are reactive under different conditions and thus may preserve different stages of the P-T evolution. Allanite, monazite, and rutile (or titanate) are well-suited chronometers for medium grade terrains, where the more reliable geochronometer zircon, is commonly not reactive (e.g. Romer et al., 1996; Vonlanthen et al., 2012, see also a review in Rubatto, 2017).

At upper greenschist-facies conditions, allanite can replace detrital or low-grade metamorphic monazite; subsequently monazite crystallises at the expense of allanite at middle amphibolite-facies conditions (e.g. Smith and Barreiro, 1990; Finger et al., 1998; Wing et al., 2003; Janots et al., 2008; Kim et al., 2009). The reactions of monazite to allanite and allanite to monazite do not take place in all rock types and at the same conditions (Engi, 2017). The proposed variables controlling the reactions include bulk composition (Finger et al., 1998; Wing et al., 2003; Janots et al., 2008; Spear, 2010), fluid composition (Budzýn et al., 2010), the oxygen fugacity of the fluid (Janots et al., 2011), and pressure (Janots et al., 2007; Spear, 2010). The use of allanite as U-Th-Pb geochronometer is complicated by internal complexity (zoning) and typically high

81 concentration of initial Pb incorporated into the mineral during crystallisation (e.g.

82 Romer and Siegesmund, 2003). The problem of initial Pb in allanite has been addressed

83 by Gregory et al. (2007) and Smye et al. (2014). It has been demonstrated that

84 regression in a Tera-Wasserburg diagram or the Th-isochron technique (Gregory et al.,

85 2007) eliminates the need to assume the isotopic composition of initial Pb and allows

86 determination of geologically significant ages even for allanite with high amounts of

87 initial Pb (Janots and Rubatto, 2014). Additional issues with allanite are the relative

88 mobility of Th and U resulting in Th-U fractionation (Smye et al., 2014). Monazite and

89 allanite potentially incorporate excess of ^{206}Pb produced by the decay of ^{230}Th, which

90 can compromise the ^{206}Pb/^{238}U ratios. Therefore, calculation of isotopic ages and their

91 interpretation for these minerals demand special attention to these complications.

92 At amphibolite facies conditions, the stable Ti-bearing phase can be rutile, titanite

93 or ilmenite; their relative stability is controlled by pressure, temperature and bulk rock

94 composition. Rutile has the advantage that it generally does not incorporate initial Pb at

95 formation, but can contain very low U concentrations that render precise measurement

96 of radiogenic isotopic ratios difficult (Kooijman et al., 2010, Warren et al., 2012; Smye et

97 al., 2014).

98 Petrography is required to link the growth of accessory minerals to metamorphic

99 structures and major mineral phases (Janots et al., 2008; Janots et al., 2011; Regis et al.,

100 2014). The relative timing of monazite and allanite can also be linked to major phases

101 such as garnet through trace element distributions (e.g. Foster et al., 2002; Hermann and

102 Rubatto 2003; Buick et al., 2006). Both allanite and monazite commonly preserve

103 multiple growth zones, and individual growth zones can be analysed by *in situ*

104 techniques. Minerals that yield ages from more than one growth zone can be used for

105   inferring metamorphic rates, provided that metamorphic conditions can be established

106   for each growth zone (e.g. Hermann and Rubatto, 2003; Janots et al., 2009; Pollington

107   and Baxter, 2010; Rubatto et al., 2011; Janots and Rubatto, 2014; Regis et al., 2014).

108   The various challenges related to monazite, allanite and rutile geochronology (e.g.

109   multiple growth zones, common and inherited initial Pb, low U content, different

110   reactivity, etc...) may reduce accuracy and precision of single age determinations. A

111   mineral-specific approach to U-Th-Pb analysis and data treatment is required because of

112   the different U-Th-Pb systematics in monazite, allanite and rutile: bulk *versus* micro-

113   sampling methods according to internal zoning, U-Pb or Th-Pb system and different

114   approaches to corrections for initial Pb. Dating of multiple mineral zones in the same

115   rock and comparison of ages from diverse lithologies, both locally and over regional

116   scale will also increase reliability of the ages.

117   We apply the multi-mineral dating approach to the classical Barrovian, middle

118   grade sequence of the Lepontine Central Alps, Switzerland. This metamorphic sequence

119   has been the subject of half a century of study, following the reaction boundary maps of

120   Niggli and Niggli (1965). Geochronology studies in the area date back four decades to

121   the pioneering work of Hunziker, Jäger, Köppel and Steiger (Hunziker, 1969; Jäger,

122   1973; Köppel and Grünenfelder, 1975; Deutsch and Steiger, 1985; Steck and Hunziker,

123   1994). Despite the long history of geochronology there remain significant uncertainties

124   and debates on the timing and duration of Barrovian metamorphism. The lack of

125   consensus is mainly due to uncertainty regarding the extent to which dated micas and

126   amphibole represent inherited, formation or cooling ages considering the metamorphic

127   grade and complex metamorphic history preserved the rocks in the Central Alps (parts

128   of which span two or more orogenic cycles). The present study is focused on areas

129   between the Northern and Southern Steep Belts, encompassing almost the entire East-

130   to-West breadth of the Lepontine dome.

131

132   **2. Geological background**

133   The Lepontine dome of the Central Alps extends between the Northern and

134   Southern Steep Belts (NSB and SSB respectively), the Simplon line in the west, and

135   Bergell area in the east (Fig. 1). The nappe stack in the dome comprises

136   polymetamorphic basement units and Mesozoic sedimentary cover units; both are

137   overprinted by a Barrovian sequence that reached maximum temperatures just north of

138   the Insubric line (e.g. Frey et al., 1974; Frey et al., 1980; Trommsdorff, 1980; Engi et al.,

139   1995; Todd and Engi, 1997). Concentric isograds and isotherms document an increase in

140   metamorphic grade from lower-amphibolite facies in the Northern Steep Belt (NSB) to

141   upper-amphibolite facies with partial melting in the Southern Steep Belt (SSB, Fig. 1b).

142   This study investigates metasediments from the cover sequences as well as

143   basement rocks (Table 1). The Permo-Mesozoic sedimentary units were deposited on

144   the subsiding European continental margin during the extensional phase in between the

145   Variscan and Alpine orogenies (Grujic and Mancktelow, 1996; Berger et al., 2005;

146   Garofalo, 2012).   During Alpine metamorphism the peak temperature was reached after

147   nappe formation and emplacement, as mineral zone boundaries and isotherms

148   discordantly crosscut tectonic boundaries (Fig. 1b; Niggli and Niggli, 1965; Frey et al.,

149   1980; Engi et al., 1995; Todd and Engi, 1997). In the southern portion of the Lepontine

150   dome, Barrovian metamorphism is considered to be syn- to post-kinematic with

151   refolding of the established nappe pile and exhumation of some high-pressure rocks

152   (Frey et al., 1980; Nagel et al., 2002; Brouwer et al., 2004; Brouwer et al., 2005; Berger et

153   al., 2005; Rütti et al., 2008; Berger et al., 2011). In the northern part of the Lepontine,

154   Barrovian metamorphism probably reached T_{max} conditions after most of the major

155   thrusts ceased to operate (Berger et al., 2005; Wiederkehr et al., 2008; Janots et al.,

156   2009; Wiederkehr et al., 2009; Berger et al., 2011). However, Wiederkehr et al. (2011)

157   showed that near the NE-margin isotherms are steeply inclined, affected by late-

158   orogenic km-scale folding, and may be displaced along a major nappe boundary (in the

159   footwall of the Adula thrust sheet). In the study area, Barrovian metamorphic conditions

160   reached temperatures of 550-650 °C and pressures of 7 kbar or less at peak temperature

161   (Engi et al., 1995; Todd and Engi, 1997).

162   The age of Barrovian metamorphism in the Central Alps is the subject of ongoing

163   debate. Pioneering studies on mica geochronology (K-Ar, Rb-Sr) from outside the

164   Lepontine dome, where peak metamorphic temperatures did not exceed the assumed

165   closure temperature of the dated minerals, led to a long dominating view that placed

166   peak Barrovian metamorphic conditions at 38-40 Ma (Jäger, 1973; Steck and Hunziker,

167   1994). On the other hand, K-Ar dating of amphiboles across the northern part of the

168   Lepontine dome returned much younger ages of 32-23 Ma that were interpreted as

169   formation ages (Deutsch and Steiger, 1985). Such younger metamorphic ages had been

170   previously documented in monazite from the centre of the Lepontine dome (Köppel and

171   Grünenfelder, 1975) but no relation of monazite age to metamorphic assemblage was

172   shown. More recent work emphasised crystallisation ages of accessory minerals, and

173   this now points to a younger age (17-32 Ma) for Barrovian metamorphism and an age

174   gradient across the belt (e.g. Janots et al., 2009; Rubatto et al., 2009; Berger et al., 2011).

175   *In situ* zircon and allanite ages for protracted melting in migmatites from the SSB span

176   from ~ 32 to 22 Ma, indicating that temperatures were high in the southernmost part of

177   the Lepontine dome up to 22 Ma (Rubatto et al., 2009; Gregory et al., 2012). Similarly,

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178   studies on rocks in the northern limits of the Lepontine dome have established an 18-19

179   Ma age for T_{max} (Janots et al., 2009). Based on U-Pb in monazite, this young age for T_{max}

180   is supported by 15-18 Ma Ar-Ar ages in white mica in rocks in the north-east of the

181   Lepontine Dome (Wiederkehr et al., 2009) and in the Lucomagno area (Allaz et al.,

182   2011).

183   The modern geochronology studies have mainly focussed on areas in the Southern-

184   and Northern Steep Belts (Gebauer 1996; Romer et al., 1996; Janots et al., 2009; Rubatto

185   et al., 2009; Wiederkehr et al., 2009), and data from the central part of the dome remain

186   scarce. In this area, zircon was largely unreactive under medium grade metamorphic

187   conditions and indeed our attempt to date the rare and thin zircon metamorphic

188   overgrowths only returned scattered dates that do not represent significant ages.

189   Systems utilising micas (e.g. K-Ar, Ar-Ar, Rb-Sr) have been interpreted to largely record

190   cooling, retrograde reactions, or they retain pre-Alpine relics (e.g. Jäger, 1973; Hurford

191   1986; Janots et al., 2009; Allaz et al., 2011).

192

193   **3. Sample description and petrography of accessory minerals**

194   Sample locations of this contribution are from as far north as Campolungo (south-

195   east of Airolo, Fig. 1b); the Western-most sample is from Croveo, north of Domodossola.

196   Eastern-most samples are from the Forcola area, $\sim$ 75 km east of Croveo and west of

197   Chiavenna. The geographical spread of samples across this central part of the Lepontine

198   dome provides ages that can reconcile the data sets of the NSB and SSB.

199   Our samples in the central Lepontine dome record sub-solidus amphibolite-facies

200   conditions during Barrovian metamorphism. For the same area, Todd and Engi (1997)

201   reported, T_{max} varying from 550°C in the lower grade rocks, to 620°C in the highest-

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202 grade samples. Pressures estimated for equilibration of the rocks (i.e. at maximum T)

203 vary between 5 kbar in the eastern-most samples and < 6.5 kbar in samples in the

204 central to western part of the study area (Todd and Engi, 1997). An overview of sample

205 locations, mineral assemblages and geochronology is presented in Table 1. Details of the

206 mineral textures and compositions are presented in the electronic Appendix. Important

207 textural relationships regarding key major and accessory minerals are shown in Fig. 2.

208 Bulk rock analyses of the samples – excluding sample Sp9312, which has distinct

209 compositional layering – are given in Table A1.

210

211 4. Mineral composition

212 4.1. Garnet

213 Croveo schist (Ma9330) garnet grains have core-rim compositional zoning (Fig. 4a,

214 Table A2). Garnet cores are rich in Mn and Ca (MnO = 5.8 wt%; CaO = 8.1 wt%), which

215 decrease towards the rims. Fe and Mg have the opposite trend, with lowest

216 concentrations in the core (FeO = 27 wt%; MgO = 0.85 wt%), increasing outwards. The

217 HREE and Y concentrations are highest in the core (Y = 370 ppm, Yb = 96 ppm) and

218 decrease dramatically before increasing again towards the outer rim. The increase in

219 HREE+Y in the outer rim is taken to indicate garnet resorption. HREE patterns are

220 steepest in the core (Dy_N/Lu_N = 0.03) and become flatter outwards (Dy_N/Lu_N ≤ 1.4).

221 Garnet outer rims are characterised by a flat to negative HREE slope (Dy_N/Lu_N > 2, Fig.

222 4b). Garnet cores and rims have a weak negative Eu anomaly of = 0.4-0.7.

223 Garnet grains in CLB-3 Campolungo calcschist show a simple core-to-rim zoning in

224 the inner part of the garnet (Grt I) with some subtle complexity in the grain rim, (Grt II,

225 Fig. 4c). CLB-3 Grt I cores are rich in Mn (MnO = 2.3 wt%) and poor in Mg (MgO = 1.6

226   wt%) compared to Grt II (MnO = 0.4 wt%; MgO = 2.4 wt%). Garnet grains in this sample

227   are remarkably low in REE (Fig. 4d, Table A2). LREE concentrations were often below

228   the limit of detection (typically 0.002 ppm for Ce, 0.003 ppm for La), and only REE

229   heavier than Eu were consistently above detection limits. HREE slopes are steep in Grt I

230   cores (Dy_N/Lu_N = 0.008) and become gentler outwards (Dy_N/Lu_N = 0.15 near Grt II). Grt

231   II is characterised by relative enrichment in some HREE, particularly Tb to Er.

232   Garnet in sample CLM-5 and 6 has a distinct core that is rich in inclusions of

233   graphite. The inclusion-poor rim is separated from the core by an irregular annulus that

234   cuts across the zoning of the core (Fig. 4e-f). The core is poor in MgO (1-4–2.5 wt%) and

235   richer in CaO (0.7-2.6 wt%) and MnO (0.7-4.2 wt%) with respect to the rim (MgO = 3.0-

236   3.5, CaO = 0.4, MnO = 0.13-0.4 wt%). The large core generally shows a decrease in these

237   elements from the inner to the outer part, and additional Mn and Ca patchy zoning in

238   CLM-5 or oscillatory zoning in CLM-6 (Fig. 4f). The core of CLM-6 garnet contains

239   inclusions of chloritoid (X_{Mg}=0.14), rare staurolite (X_{Mg}=0.07), chlorite, quartz, ilmenite

240   and rutile. The discordant rim is more homogeneous in composition with a slight

241   increase in Mg outwards. Ilmenite, rutile, chlorite, quartz and rare chloritoid (X_{Mg}=0.19)

242   inclusions have been observed. Staurolite in the matrix coexists with garnet rims and

243   displays significantly higher X_{Mg}=0.21 than staurolite inclusions.

244   Alpe Sponda fels Sp9712 contains garnet with complex zoning in major and trace

245   element composition. Because garnet grains are scarce in Sp9712, they were analysed as

246   polished grains retrieved from the sample separate. Ca element maps of larger grains

247   show an outward increase in Ca from the core (Grt I), before an abrupt drop in

248   concentration in Grt II (Fig. 4g). A number of garnet grains show embayment to the Ca

249   zoning. In these grains, Fe shows a complementary trend to Ca, decreasing from the core

11

250   before increasing in Grt II. The trace element zoning is best seen in HREE

251   concentrations. Grt I cores display little zoning (Fig. 4g) and have a flat HREE pattern.

252   Towards the rim of Grt I the HREE significantly decrease. Grt II is marked by an increase

253   in HREE to higher levels than what is observed in Grt I cores, followed by a decrease

254   towards the rims of Grt II.

255   One small (200 μm diameter) euhedral garnet grain with distinctive composition

256   (Grt III) presents a relatively simple, bell-shaped zoning with cores enriched in Ca and Y,

257   which decrease towards the rims. HREE are even more extremely enriched than in Grt II

258   (Dy_N/Lu_N = 0.1-0.4, Fig. 4h) with greatest HREE enrichment occurring in the core. Y

259   concentration is as high as 3 wt % in garnet cores and decreases to ∼ 3500 ppm at the

260   rim. Element maps of major and trace elements suggest that Grt II and Grt III formed at

261   the same time, with the first one as overgrowth on pre-existing, partially resorbed cores

262   and the latter as newly formed, small individual grains.

263   *4.2. Allanite*

264   *Metasediments*. The compositional zoning of allanite is similar in the metasediment

265   samples from the western and central areas (Robiei Ba0901, Campolungo CLB-3 and

266   Ma0901); they are considered here together. Allanite has three distinctive zones: a

267   LREE-enriched core, a HREE-enriched mantle, and an epidote rim (Fig. 3a-d). Ba0901

268   has one additional allanite rim that is overgrown on epidote.

269   Robiei (Ba0901) LREE-allanite cores and rims have similar REE patterns (Fig. 5a)

270   with a significant negative Eu anomaly (Eu/Eu* = 0.3-0.5 in cores and 0.2-0.5 in rims)

271   and a similar HREE slope (Dy_N/Lu_N = 4.5 in cores and 5.5 in rims). Ba0901 allanite cores

272   vary in Th from 4300 to 12000 ppm, U spans from 600 to 1500 ppm, and Th/U ratio

273   varies from 2.8 to 12 (Table A4). Ba0901 LREE-allanite rims are similar in composition

274   to cores of the same grains but differ in the abundance of HREE, which tend to be higher

275   in the rims than cores. Th concentration is lower in rims (2000-8000 ppm) and U

276   concentration is similar to cores (300-1700 ppm). Th/U ratios show little variation, with

277   Th/U between 3.5 and 6.5 in allanite rims.

278   Samples from Campolungo (CLB-3 calcschist and Ma0901 quartzite, Fig. 3c-d) both

279   lack the LREE-rich rim characteristic of the Robiei (Ba0901) allanite. The REE patterns

280   of allanite cores in both samples have a significant negative Eu anomaly (Eu/Eu* = 0.6 in

281   CLB-3 and 0.5 in Ma0901) and a steep HREE pattern (CLB-3 Dy_N/Lu_N = 7-17, Ma0901

282   Dy_N/Lu_N = 8-24; Fig. 5b-c). CLB-3 calcschist allanite cores have lower Th and U

283   concentrations than in the quartzite (Ma0901). CLB-3 allanite cores contain 4300-7700

284   ppm Th and 100-400 ppm U; Ma0901 allanite contains 3300-18000 ppm Th and 400-

285   1200 ppm U. Th/U ratios in CLB-3 are high and vary from 40 to 77; Th/U ratios in

286   Ma0901 quartzite vary from 2.8 to 27.

287   Allanite mantles with a relative HREE- and U-enrichment (Fig. 5d) are only found

288   in the metasediments. The HREE-allanite mantles are commonly thin and can have

289   complex internal zoning, especially in the case of Ba0901 (Fig. 3a-b). In all allanite

290   mantle zones, HREE concentrations decrease outwards. Campolungo allanite from both

291   CLB-3 calcschist and Ma0901 quartzite has significant HREE enrichment in the mantle

292   zone, and the HREE pattern has a concave down shape (Fig. 5b-c). HREE-allanite from all

293   samples shows a similarly weak negative Eu-anomaly (Eu/Eu* ≈ 0.75). U concentrations

294   are much lower in the Campolungo calcschist than the quartzite (500 to 2300 ppm in

295   CLB-3 calcschist and 1200 to 3500 ppm U in Ma0901); Th/U ratios in mantle allanite are

296   much lower than in the cores for both samples (CLB-3 Th/U = 2-3; Ma0901 Th/U = 0.4-

297   1).

298   *Forcola orthogneiss*. Despite the great variety of internal textures (Fig. 3f-h),

299 allanites from orthogneiss samples (LEP0979, LEP0980, LEP0807) have similar trace

300 element compositions. For all Forcola orthogneiss allanites, REE patterns have

301 significant negative Eu anomalies (Eu/Eu* = 0.1-0.6, Fig. 5e-f) and a comparable HREE

302 slope with Dy_N/Lu_N varying from 2 to 18. Th concentrations in orthogneiss allanites vary

303 from 2000 to 20000 ppm; U concentration spans from 400 to 1200 ppm. The highest U

304 concentrations occur in patchy allanite in sample LEP0807. Th/U ratios typically span

305 between 4 and 31 in LEP0979 and LEP0807 allanites. LEP0980 regularly zoned allanite

306 of Alpine age have a much lower Th/U ratio (Th/U = 0.2 to 12). The spectacularly zoned

307 LEP0980 mosaic allanite have a trace element composition that is not distinctive and is

308 within the range of the other Forcola orthogneiss allanites.

309 *5.4. Monazite*

310   REE composition of monazite is very similar in both samples (Croveo schist

311 Ma9330, Alpe Sponda fels Sp9712) with a negative Eu anomaly (Eu/Eu* 0.5– 0.4) and a

312 variable HREE slope (Fig. 5g, Table A5). U concentrations are similar in both samples

313 (Ma9330 U = 3900-6300 ppm; Sp9712 U = 5400-6600 ppm) and Th concentrations are

314 all but identical (Ma9330 Th = 21.8-34.1 %; Sp9712 Th = 25.2-34.7 %). Th/U ratios tend

315 to be higher and more variable in Ma9330 monazite than in that from Sp9712 (Ma9330

316 Th/U = 5.0-7.9; Sp9712 Th/U = 4.6-6.3).

317

318 **5. Rutile geothermometry**

319   Selected rutiles were analysed for trace elements using LA ICP-MS (Table A3). The

320 Zr content of rutile is a geothermometer (Ferry and Watson, 2007) that can provide

321 crystallisation temperature estimates for the rutiles that are related to the growth of

14

322 allanite cores (and hence their age) in Ma0901 quartzite and to the rutile bearing-

323 paragenesis and/or ages in samples Ma9330, Ba0903A, CLB-3, CLB-4 and Sp9712.

324 Following Ewing et al. (2013), the sources of uncertainty to be propagated onto Zr-in-

325 rutile temperature estimates are the effect of analytical uncertainty of Zr measurement

326 and uncertainty inherent in the calibration (~3 % for 500 °C, Ferry and Watson, 2007).

327 The Zr analytical error was conservatively estimated to be ± 15 %, which corresponds to

328 an error of ± 10-15 °C for the 500-650 °C temperature range and combined with the

329 internal error of the calibration of Ferry and Watson (2007) it gives a total uncertainty

330 of ± 20 °C for these samples.

331   Rutile from sample Ma9330, the western-most sample, returns a Zr-in-rutile

332 temperature of ~ 565 °C (Zr = 100 ppm), clearly lower than T_{max} of 620 °C from Todd

333 and Engi (1997). Ba0903A is of significantly lower grade than Ma9330 (T_{max} for this area

334 is 575 °C, Todd and Engi, 1997) and the range of Zr concentrations in rutile from 100 to

335 166 ppm, corresponds to 565-595 °C. Samples CLB-3, CLB-4 and Ma0901 are all from

336 the same locality and return Zr-in-rutile temperature estimates of ~ 565 °C, 625 °C and

337 580 °C, respectively (Zr = 100, 240 and 130 ppm, respectively). Sp9712 is the eastern-

338 most rutile-bearing sample and returns a Zr-in-rutile temperature estimate of 550 °C (Zr

339 = 85 ppm), which is again below the T_{max} expected for this area according to Todd and

340 Engi (1997).

341

342 **6. Geochronology**

343   Depending on the assemblages of the investigated samples, monazite, allanite or

344 rutile were dated. These minerals have different Th-U contents (rutile is virtually Th free

345 and contains only 0.3 to 20 ppm U, monazite has Th/U of 5–10 and Alpine allanite has

346 Th/U of 2–80) and can variably incorporate initial Pb. Additionally, rutile shows no

347 evident internal zoning, monazite shows limited evidence of internal zoning, whereas

348 allanite is strongly zoned in both composition and age. Because of these inherent

349 differences, geochronology of these minerals followed different approaches to achieve

350 best age accuracy and precision.

351   Rutile was dated by ID-TIMS because of its low U content and the lack of intragrain

352 zoning. Due to low U content, the measured Pb composition has a low radiogenic

353 component and a significant proportion of initial Pb (Supplementary Table A8). Because

354 these are multigrain fractions from different samples, with apparent different age they

355 cannot be used for an isochron calculation and the best way to determine the initial Pb

356 composition for each aliquot was to measure the composition of low-U-feldspar (Table

357 A8b).

358   In the investigated samples, allanite is rich in Th and U, and both systems are

359 strongly contaminated by initial Pb. The Th-Pb system in these samples is relatively

360 more radiogenic than U-Pb (Supplementary Table A7) and thus it was preferred for age

361 determination. Measuring the Pb composition of feldspar is not a rigorous approach to

362 estimate the initial Pb composition for allanite in these samples because of the zoning

363 complexity, the presence of different age populations, and the possibility of inherited

364 initial Pb from precursor monazite (e.g. Romer and Rötzler, 2011). A better approach is

365 to calculate Th-isochrons according to the methods of Gregory et al. (2007; $^{208}Pb/^{206}Pb_c$

366 versus $^{232}Th/^{206}Pb_c$ where $^{206}Pb_c$ is initial Pb only) for multiple analyses of the same

367 domain in each sample. In this approach the assumption on common Pb composition

368 (adopted to be the model composition of Stacey and Kramers 1975) has only marginal

369 bearings on the calculation of the $^{206}Pb_c$ (see Gregory et al., 2007; Janots and Rubatto,

370 2014).

371   In monazite, unlike allanite, the initial Pb incorporation is low (mostly <3%) and

372   thus the assumption on the common Pb composition has no bearing on the age within

373   uncertainty. The U-Pb system was preferred to the Th-Pb system because of analytical

374   setup (see methods).

375   *6.1. Western Samples: Croveo (Ma9330) and Robiei (Ba0901)*

376   Alpine ages are summarised in Fig. 6 and data are presented in Tables A5-A7.

377   Monazite is the only accessory phase dated in the western-most sample Ma9330. The

378   monazite grains are unzoned in BSE images and return a $^{206}Pb/^{238}U$ age of 21.7 ± 0.4 Ma

379   (95 % confidence for all average ages, Fig. 7b) that is within error of the monazite age

380   from central sample Sp9712 (see below).

381   Accessory allanite was dated in the Robiei sample Ba0901. Pre-Alpine igneous

382   allanite relics returned dates that span from 397 to 187 Ma, a spread that may indicate

383   alteration or partial resetting of the $^{232}Th-^{208}Pb$ isotopic system. Metamorphic allanite

384   cores return an isochron age of 26.8 ± 2.3 Ma (MSWD = 2.0, n = 9/10; Fig. 8a). The

385   scatter in the data is likely to be due to some minimal overlap of the SHRIMP analysis

386   spot onto the surrounding growth zones, because of the small size of the cores. Allanite

387   rim analyses define an isochron age of 19.7 ± 1.3 Ma (MSWD = 0.77, n = 11/12; Fig. 8b).

388   Rutile from Ba0903A calcschist returns a range of dates for different fractions that

389   scatter from 21 to 27 Ma (Table A8). The oldest rutile date is from fraction A45 40, at

390   27.4 ± 1.7 Ma, which is similar to that of allanite cores from the same locality (Ba0901).

391   Two fractions of orange grain fragments returned dates of 21.5 ± 2.5 Ma and 26.1 ±2.2

392   Ma.

393   *6.2. Central samples: Campolungo (CLM-5, CLM-6, CLB-3, CLB-4, Ma0901) and Alpe Sponda*

394   *(Sp9712)*

395   Ages from allanite and monazite were extracted from the central samples (Fig. 6).

396   Monazite in sample CLM-5 and 6 is pre-Alpine in age despite being aligned along the

397   main foliation. They have weak internal zoning with sector and flame texture. Both

398   samples returned a majority of dates between 322 and 339 ± 4 Ma (1σ) that define

399   average ages of 331 ± 4 Ma and 326 ± 4, respectively (Fig. 7a). In both samples a few

400   analyses show scattering towards younger dates that likely reflect Pb disturbance

401   during Alpine overprint.

402   Allanite from CLB-3 calcschist returns an isochron age of 31.7 ± 1.1 Ma (MSWD =

403   1.5, n = 15/17, Fig. 8c), which is older than Ma0901 allanite cores from the same locality

404   (28.5 ± 1.3 Ma, MSWD = 0.60, n = 27/29, Fig. 8d). Notably, for both samples, the

405   weighted means of the most significant cluster of single spot dates – corrected for Pb_c

406   using the model common Pb composition of Stacey and Kramers (1975) – are within

407   error of the isochron ages (CLB-3 mean age 31.8 ± 0.9 Ma, MSWD = 1.7, N 10; Ma0901

408   29.6 ± 0.5 Ma, MSWD = 1.2, N 16). However, for the reasons given by Gregory et al.

409   (2007) and Janots and Rubatto (2014), the isochron ages are deemed more reliable and

410   are used in the discussion. Ma0901 HREE-rich allanite mantles are large enough to be

411   analysed by SHRIMP but contained prohibitively high ƒ208 (> 90%) and the data do not

412   form a reliable isochron, preventing a reliable age calculation.

413   Rutile dates from one Campolungo sample (CLB-4) span from 19 to 25 Ma (Table

414   A8). Like Ba0903A rutiles, the fractions comprising dark-coloured and larger grains and

415   fragments tend to be older (~ 24 Ma, 4 fractions; Table A8) than the lighter honey-

416   coloured rutile grains (~ 21 Ma, 3 fractions).

417   Sp9712 monazite returns a ^{206}Pb/^{238}U age of 22.0 ± 0.3 Ma (Fig. 7c; Table A6).

418   Monazite grains are subtly heterogeneous in BSE images (Fig. 7c), but there is no

419   correlation between zoning and age. The monazite age is within error of the monazite

420   age from Ma9330. Two groups of dates were obtained from Sp9712 rutile (Table A8).

421   Dark red-brown rutiles returned older dates between 52 and 57 Ma; the apparently

422   younger rutiles are orange and light honey-coloured grains at 33.45 ± 0.37 Ma (Table

423   A8).

424   *6.3. Eastern samples: Forcola Orthogneiss (LEP0979, LEP0980, LEP0807)*

425   Two orthogneiss samples contain metamorphic allanite that return acceptable

426   isochron ages (MSWD < 2). Subgrain cores from LEP0980 mosaic allanite return an

427   isochron age of 31.3 ± 1.1 Ma (MSWD = 1.1, n = 16/16; Fig. 8e); allanite from LEP0807,

428   the structurally shallowest Forcola sample, return an isochron age of 27.4 ± 0.6 Ma

429   (MSWD = 1.9, n = 17/21; Fig. 8f).

430   Allanite samples that did not form meaningful isochrons (MSWD > 2) are LEP0979

431   allanite, regularly zoned LEP0980 allanite, and subgrain rims of LEP0980 mosaic

432   allanite. For these sample ^{207}Pb-corrected single spot dates are presented in Table A7,

433   and in all cases they show a range of several Ma, suggesting the isotopic systematic was

434   disturbed or individual grains contained variable initial Pb. Such dates are not taken

435   individually to constrain significant geological events.

436

437   **7. Discussion**

438   *7.1. Reliability of ages*

439   At low to medium metamorphic grade allanite and monazite can replace each

440   other (Janots et al., 2008; Janots et al., 2009) or grow from previous magmatic allanite or

441   monazite with possible incorporation of radiogenic Pb from the precursor phase (e.g.

442   Romer and Siegesmund, 2003; Romer and Rötzler, 2011). This inheritance has bearings

443   on initial Pb composition and thus age calculation. For the analysed monazite the

444   fraction of initial Pb is small enough to make this problem minimal. Any incorporation of

445   inherited radiogenic Pb would imply a lower $^{207}Pb/^{206}Pb$ ratio of the initial Pb and

446   mildly shift the calculated age to younger values. Notably any excess ^{206}Pb may lead to a

447   shift to older ages (see below).

448   For allanite the presence of inherited radiogenic and common Pb is a more serious

449   problem, but the Th-isochron calculation is largely independent from the initial Pb

450   composition, which is only used to determine the fraction of initial ^{206}Pb (see Gregory et

451   al., 2007). Additionally, in the sample investigated, single spot ages calculated assuming

452   a secular Pb composition (Stacey and Kramers, 1975) are mostly close to the Th-

453   isochron age, indicating that initial Pb incorporated at growth was generally non

454   radiogenic. This observation also limits the possibility of a shift in the age due to

455   chemical inheritance as described by Romer and Siegesmund (2003) and Romer and

456   Rötzler (2011). For allanite domains that do not yield an isochron age, the range of

457   single spot dates are not given a geological significance as they may be affected by Pb

458   inheritance. This is likely the case for the mosaic allanite in orthogneiss LEP0980 (Fig.

459   3h), where the subsolidus reaction that recrystallized the pre-Alpine allanite into the

460   mosaic Alpine allanite was likely a closed system reaction and thus the Pb did not

461   equilibrate.

462   Excess ^{206}Pb due to the incorporation of ^{230}Th at growth is another issue to

463   consider in dating Th-rich minerals. Unlike magmatic systems, in metamorphic rocks a

464   correction for excess ^{206}Pb based on the Th/U of the system is not possible because of

465   changing reactive bulk during metamorphism. While this does not affect the Th-Pb

466   system used for allanite dating, it can produce apparently older $^{206}Pb/^{238}U$ ages for

467   monazite. Unlike magmatic monazite grown from a melt, excess ^{206}Pb in metamorphic

468   monazite has been rarely reported (see Rubatto et al., 2013; Wang et al., 2015).

469   Monazite analyses in this study are concordant within analytical uncertainty and this

470   indicates that any excess ^{206}Pb is less than the analytical uncertainty of the $^{206}Pb/^{238}U$

471   ratio of single analyses, that is 0.6–1.5% for Variscan monazite and 2–4% in Alpine

472   monazite.

473   In all analysed samples, rutile U contents are low, (mostly below 15 ppm with the

474   exclusion of sample CLB where U content is 26-36 ppm, Table A3) and the proportion of

475   initial Pb is thus high, with molar fraction of non-radiogenic ^{206}Pb between 0.75 and 0.98

476   (Table A8). To attempt non-radiogenic Pb correction, the Pb composition of K-feldspar

477   was measured in sample CLB4 and Sp9712, but a precise estimate was only obtained for

478   Sp9712. Using this feldspar Pb composition to correct the data results in dates scattering

479   from ~ 20 to 54 Ma. However, particularly the dates above 23 Ma have a positive

480   correlation to the fraction of non-radiogenic Pb, an indication that the chosen initial Pb

481   isotopic composition may be not representative for the rutile. Applying this initial Pb

482   correction, the resulting rutile dates are mostly Alpine with a suspected pre-Alpine

483   component in the Alpe Sponda basement (see further discussion below). The U-Pb

484   analyses do not form a well defined regression in an uncorrected Tera Wasserburg plots,

485   and thus the composition of the initial Pb could not be calculated by this method. Pb-Pb

486   isochrons also failed due to data scatter. Because of the low Pb radiogenic component in

487   the rutile and the issue with initial Pb correction, the rutile U-Pb dates should be used

488   and interpreted with extreme caution. Whenever possible, we prefer to rely on more

489   robust age data from other mineral chronometers

490

21

7.2. Conditions of allanite, monazite and rutile formation

Constraining the condition of formation of allanite and monazite with respect to structures and metamorphism is crucial for age interpretation. During Alpine metamorphism, the peak temperatures were below 650°C in all investigated samples (Fig. 1), which is well below the closure temperature for the Th-U-Pb system in allanite (Gregory et al., 2012) and monazite (Rubatto et al., 2001). This is further confirmed for allanite by the presence of inherited ages in the Forcola orthogneisses and the Robiei gneiss Ba0901. Therefore, the obtained ages date the formation of the mineral growth zones. We use textural relationships and the trace element composition of monazite and allanite growth zones to constrain the reactions that took place during allanite-epidote growth and infer coexistence of accessory minerals and major phases such as garnet (e.g. Rubatto, 2002; Hermann and Rubatto, 2003; Buick et al., 2006). Zr-in-rutile thermometry can be used to relate rutile ages to metamorphic conditions. However, in the present case, age interpretation is complicated by the potential mixing of different rutile generations when 100s of grains are picked for isotope dilution analysis and the fact that Pb diffusion in rutile may occur at amphibolite-facies conditions over the relatively wide range of ~450 to 650 °C (e.g. Cherniak, 2000; Vry and Baker, 2006; Kooijman et al., 2010; Warren et al., 2012). Below we discuss the ages from across the area in chronological order, starting from Variscan metamorphic ages in the basement before discussing prograde Alpine allanite to higher temperature monazite.

Pre-Alpine monazite in Campolungo schists CLM-5 and 6 is aligned along the folded main foliation (Fig. 2e), which is partly wrapping the garnet, but is overgrown by staurolite and kyanite. In both samples monazite contains black micro inclusions of graphite, which occasionally defines a foliation that continues in the matrix (Fig 2c). This suggests that the Carboniferous monazite already formed as part of a metamorphic

516 assemblage. The crystals are euhedral, flat prismatic in shape, with sharp edges and

517 yield a single age and therefore a detrital origin is excluded. The ~330 Ma age in the two

518 samples is interpreted as dating Variscan metamorphism in the Campolungo (Simano

519 nappe) basement. It is surprising that no Alpine monazite has been found in both

520 samples. Garnet in CLM-6 displays a core with concentric zoning that is separated from a

521 nearly homogenous rim by an irregularly shaped annulus that is crosscutting the core. In

522 the light of a pre-Alpine age of metamorphic monazite, this feature is interpreted as a

523 phase of garnet resorption between the Variscan metamorphic event and the Alpine

524 amphibolite-facies metamorphism. Accordingly, the Variscan metamorphic conditions

525 can be constrained to be of upper greenschist to lower amphibolite facies based on the

526 inclusion assemblage of chloritoid, chlorite and staurolite found in the garnet core. At

527 these general conditions, rutile may have been the stable Ti phase. Formation of rutile

528 during a Variscan event is suggested on the basis of the high apparent ages obtained in

529 Alpe Sponda fels Sp9712, where two fractions containing darker rutile grains returned

530 dates of ~ 52 to 57 Ma. Such spurious dates could be due to mixing between Alpine and

531 Variscan rutile, possibly with partial Pb loss of the older population.

532   The pre-Alpine metamorphic event was overprinted during Alpine metamorphism

533 by the amphibolite-facies assemblage of muscovite-biotite-garnet-staurolite at only

534 slightly higher metamorphic conditions. The main foliation S1 in schists CLM-5 and 6,

535 that is parallel to the main foliation in the overlaying metamorphosed Mesozoic

536 sediments, is therefore interpreted as a transposed pre-Alpine foliation. Gieré et al.

537 (2011) reported very similar garnet and tourmaline textures with a Ca-rich annulus

538 separating cores and rims from metapelites from the same area. As the protolith was

539 assumed to be a Permo-Carbonferous sediment (and thus would have experienced only

540 Alpine metamorphism), they interpreted that the annulus was formed during Alpine

541 prograde metamorphism at the transition from allanite to monazite. This is inconsistent

542 with the new age data that shows that the rocks around Laghetto di Campolungo belong

543 to the polymetamorphic basement of the Simano nappe and that no new monazite was

544 formed by the Alpine metamorphic cycle. Indeed, the reported trace element

545 compositions of monazite reported by Gieré et al. (2011) include 1200 ppm Pb, 42000

546 ppm Th and 5100 ppm U. The high amount of Pb is consistent with a Variscan age of the

547 monazite rather than an Alpine age (that would result in at least an order of magnitude

548 lower Pb content). Peak metamorphic temperatures for the Campolungo area have been

549 constrained between 600 and 625°C based on multi-phase equilibria (Fig. 1; Todd and

550 Engi 1997). On the other hand, Gieré et al. (2011) proposed a significantly higher peak T

551 of 660°C concomitant with small amounts of partial melting. Partial melting is well

552 known to enhance the recrystallization of monazite (Rubatto et al., 2001; Kelsey et al.,

553 2008). The absence of leucosomes in the field and the lack of Alpine age monazite in

554 these rocks are in better agreement with the lower peak metamorphic temperatures

555 proposed by Todd and Engi (1997).

556   Early Alpine dates around 33-34 Ma were obtained from the two fractions of Alpe

557 Sponda rutiles. In this sample, rutile is included both in large garnet grains (Grt I, II) and

558 small garnet grains (Grt III). Zr-in-rutile thermometry returns ∼ 550 °C for all Sp9712

559 rutile grains analysed (most likely including both > 50 Ma and 33-34 Ma rutile), which is

560 significantly lower than the expected T_{max} for this area (∼620 °C, Todd and Engi, 1997).

561 This age has to be taken with caution, due to potential mixing with inherited

562 components, but it is possible, and in line with allanite data, that 33-34 Ma rutile formed

563 during a prograde stage at temperatures of ∼ 550 °C.

564   The Alpine ages in the Campolungo area are constrained by accessory phases

565   found in the metamorphosed Permo-Mesozoic sedimentary rocks. Allanite in the

566   calcschist CLB-3 (31.7 ± 1.1 Ma) is included in Grt I and is aligned along the relic fabric

567   preserved in garnet (S1, Fig. 2c); garnet grains are wrapped and rotated by the

568   dominant foliation (S2). The allanite age is therefore interpreted to date prograde garnet

569   growth as well as the relic foliation along which the allanite is aligned. The dominant

570   foliation in this sample (S2) is correlated with the regional D2 (Grujic and Mancktelow,

571   1996; Maxelon and Mancktelow, 2005), the major deformation phase that refolded the

572   nappe stack and is responsible for many of the major tectonic structures in the

573   Lepontine dome. The relic foliation that is only preserved in garnet is likely to be S1,

574   and the allanite age (31.7 ± 1.1 Ma) may reflect the timing of the regional D1 nappe

575   stacking (Maxelon and Mancktelow, 2005). The HREE patterns in CLB-3 allanite cores

576   are depleted (Fig. 5b), providing evidence that allanite cores grew during/after prograde

577   garnet formation, which sequestered Mn and HREE.

578   Allanite in Campolungo quartzite Ma0901 (28.5 ± 1.3 Ma) is aligned along the

579   dominant foliation of the rock, which is likely to be a composite S1/S2 fabric (S1 and S2

580   are approximately parallel; e.g. Maxelon and Mancktelow, 2005). Rutile is included in

581   Ma0901 allanite cores and Zr-in-rutile thermometry indicates a temperature of ~

582   575 °C, which is below the estimated temperature for T_{max} conditions (600-625°C, Todd

583   and Engi, 1997). The allanite age is therefore interpreted to date a stage in the prograde

584   history of the rock during major tectonic reworking of the nappe pile (D2, Grujic and

585   Mancktelow, 1996; Maxelon and Mancktelow, 2005). Although garnet is not abundant in

586   the sample, the HREE depletion of allanite provides evidence that garnet was present at

587   the time of allanite formation (Fig. 5c).

588   The presence of relic igneous allanite in the Forcola orthogneisses and the Robiei

589   gneiss Ba0901 (the bulk composition of which is similar to orthogneiss, Table A1)

590   indicates that, in these samples, igneous allanite is the precursor to metamorphic

591   allanite. In the Robiei gneiss (Ba0901), metamorphic allanite cores (26.8 ± 1.3 Ma)

592   formed during prograde metamorphism and contain inclusions of biotite and muscovite.

593   Allanite domains in this sample show only a small decrease in total REE concentration

594   from cores through mantles and rims. It is speculated that allanite rims (19.7 ± 1.3 Ma)

595   formed by resorption whereby the allanite was partially consumed and recrystallised as

596   LREE allanite rims near T_{max} conditions. This resorption likely occurred with very little

597   volume change (i.e. REE in allanite were neither concentrated nor diluted during

598   resorption). In this sample K-feldspar and plagioclase are fresh, indicating minimal fluid

599   influx during retrogression.

600   Rutile from Robiei calcschist (Ba0903A; Bündnerschiefer) returns TIMS dates that

601   span from 27 to 21 Ma – almost the entire range of allanite and monazite ages in the

602   western samples. Zr-in-rutile thermometry indicates that rutile crystallised over a

603   limited temperature range from 565 to 595 °C. The Zr-in-rutile temperatures overlap

604   with the expected T_{max} for this area, based on multiphase equilibria thermobarometry

605   (Figure 1; Todd and Engi, 1997). Galli et al. (2007) investigated in detail the structures

606   and metamorphism of Bündnerschiefer in the Naret region, which is situated 5 km to the

607   northeast of Robiei. They proposed that main recrystallization of minerals (including

608   rutile) occurred between D2 and D3 and T_{max} of 650 °C. The range of rutile dates is

609   consistent with either several episodes of rutile recrystallization during the D1-D3

610   deformation events or partial Pb loss from an early (>27 Ma) rutile generation formed

611   during prograde metamorphism.

612   Forcola orthogneiss allanite (scattering dates 25-32 Ma) is aligned along S1, a

613   dominant and early foliation. However, the timing of allanite crystallisation with respect

614   to metamorphic phase and deformation history is complicated by the large variety of

615   internal textures of allanite (Fig. 3f-h). The mosaic allanite from LEP0980 has a unique

616   texture of subgrain cores and rims, which reflect two allanite growth stages that

617   occurred at 31.3 ± 1.1 Ma for the cores and younger in the rims. Analyses from the

618   regularly zoned allanite from the same sample do not define an isochron age and single

619   spot dates span the entire range of mosaic allanite ages (25-32 Ma). There is no

620   correlation between the age of regularly zoned allanite and internal texture (i.e. core vs.

621   rim). It is possible that the older dates are the result of Pb inherited from a precursor

622   phase (Romer and Siegesmund, 2003), and thus these ages are not further considered in

623   the discussion. In allanite from sample LEP0807 there is no correlation between Th-Pb

624   age and internal texture (core vs. rim) and all analyses defined a Th-isochron age of 27.4

625   ± 0.6 Ma suggesting that any difference in age between allanite core and rim is below the

626   resolution of the analyses.

627   Croveo schist (Ma9330) monazite age of 21.7 ± 0.3 Ma is interpreted to date the

628   timing of monazite crystallisation at the expense of allanite. Allanite as a precursor to

629   monazite (see also Romer and Siegesmund, 2003; Janots et al., 2008) is inferred from

630   the presence of relic allanite included in garnet. The HREE pattern of monazite is

631   depleted with respect to the bulk rock, suggesting that garnet was still stable when

632   monazite was forming (Fig. 5h). Additionally, the negative Eu anomaly is more

633   pronounced in monazite than in the bulk rock and thus plagioclase was also stable. As

634   garnet and plagioclase are peak metamorphic minerals, it is reasonable to assume that

635   monazite formed close to T_{max} conditions of 620°C at 6.2 kbar (Todd and Engi, 1997).

27

636   The paragneisses of Alpe Sponda belong to the basement of the Simano nappe and

637   thus it is plausible that these samples underwent polymetamorphism as is the case for

638   the Campolungo metapelites (see above). Element maps as well as trace element profiles

639   through the large garnet grains suggest that Grt I has been resorbed prior to the

640   formation of Grt II rims and newly formed Grt III. This is very similar to the two

641   generations of garnet found in sample CLM-6, where a pre-Alpine garnet core is

642   truncated by an Alpine rim. Two distinct types of garnet with the same compositional

643   characteristics as in our study (Fig. 4g-h) have been reported by Beitter et al. (2008)

644   from the Alpe Sponda area. Beitter et al. (2008) report garnet grains corresponding to

645   Grt I were only found in the paragneiss whereas grains corresponding to Grt III were

646   associated to the Alpine rutile-kyanite-quartz veins.

647   Allanite is a precursor to monazite in Alpe Sponda fels Sp9712. Rare allanite has

648   been observed in the garnet whereas no monazite is observed as inclusions in garnet.

649   The assembly of small monazite grains that satellite relic allanite (Fig. 3e) also indicates

650   that monazite replaced allanite in these rocks. The Alpe Sponda fels monazite (22.0 ± 0.3

651   Ma) returned the youngest ages of the Central samples. The steady increase of Dy/Lu

652   and an associated increase of the negative Eu anomaly (Fig. 5h) suggest that this

653   monazite formed together with garnet and plagioclase, the peak assemblage. Thus, we

654   conclude that peak metamorphic conditions persisted up to 22 Ma in this area.

655   Campolungo calcschist (CLB-4) returned rutile dates between 19-25 Ma. Rutile

656   crystallised after garnet (one of the earliest prograde minerals preserved) and before

657   staurolite (one of the later-stage minerals). The Zr-in-rutile temperature for CLB-4 is ~

658   625 °C, similar to the estimate for maximum Barrovian temperature conditions in this

659   area (600-620 °C; Todd and Engi, 1997). The spread of rutile dates may again reflect

660   protracted rutile crystallisation during slow heating while temperatures approached

661   T_{max} and/or cooling ages.

662   *7.3. Implications for the allanite-to-monazite transition in metamorphic rocks*

663   Our regional scale data set provides an excellent framework to shed light on the

664   nature of the allanite-to-monazite transition during Barrovian metamorphism (Fig. 9).

665   Rutile inclusions in allanite from Campolungo Ma0901 quartzite provide evidence that

666   the crystallisation of allanite cores occurred at a temperature of ~ 575 °C, as determined

667   by Zr-in-rutile thermometry of rutile inclusions. Moreover, the similarity of the allanite

668   ages from the calcschist and quartzite (~ 32 Ma and ~ 28 Ma respectively) and their

669   textural positions associated with the amphibolite-facies S1 and S2 foliation, indicates

670   that allanite crystallisation in the calcschist did not occur at the low temperatures of

671   400-450 °C suggested by previous studies of prograde allanite (e.g. Smith and Berreiro,

672   1990; Wing et al., 2003; Janots et al., 2008; Rasmussen and Muhling, 2009; Janots et al.,

673   2011).

674   Robiei gneiss (Ba0901) allanite is an example of two separate phases of allanite

675   crystallisation in the same rock under different conditions. Allanite prevails in this rock,

676   which contains very little Ca (0.74 %), and has a low Ca/Al ratio of 0.08. At the

677   metamorphic conditions attained and for a rock of this bulk composition, the models of

678   both Spear (2010) and Wing et al. (2003) predict monazite stability (Fig. 9). It is inferred

679   that allanite rims crystallised (instead of monazite) at the expense of allanite because

680   T_{max} was too low for monazite stability in this sample (T_{max} ~ 575 °C).

681   Monazite crystallisation in samples Ma9330 and Sp9712 is interpreted to have

682   occurred close to T_{max} conditions (~ 620 °C). This is somewhat higher than the reaction

683   temperatures suggested by previous studies of natural samples (560-580 °C in Janots et

684   al., 2008; ~ 540 °C in Gieré et al., 2011) and in thermodynamic models (Spear 2010).

685   The model of Spear (2010) proposes a temperature range of 400 to 700 °C for the

686   allanite-to-monazite reaction, depending on bulk Ca, Al and pressure (Fig. 9a,b). For bulk

687   composition of sample Ma9330 (Table A1) the model predicts that the allanite-to-

688   monazite reaction would occur at ~ 450 °C (P = 5 kbar); at 10 kbar the reaction is

689   predicted to occur at ~ 500 °C. However the model assumes simple systems and the

690   studied samples are more complex; neither of the reaction temperature predictions is

691   consistent with our observations. Furthermore, the model predicts that the reaction

692   would have occurred even in samples where allanite is the stable REE mineral (Fig.

693   9a,b). Spear and Pyle (2010) admit that good thermodynamic data for allanite is lacking

694   and the disagreement between observations in this study and predicted reaction

695   temperatures may be due to the thermodynamic data used. One of the major limitations

696   in applying thermodynamic models to accessory mineral reactions is the difficulty in

697   defining the reactive bulk composition. Particularly for minerals whose stabilities are

698   controlled by trace elements, their reactive bulk is likely not equivalent to the rock

699   composition and local difference in trace element distributions will have major

700   consequences on accessory mineral reactions. Additionally, deformation and partial

701   hydration may localize reactions. The survival of Carboniferous monazite in

702   Campolungo samples CLM-5 and 6 provides evidence that in these basement samples no

703   Alpine prograde allanite formed. This might be related to a limited

704   retrogression/hydration of these amphibolite-facies schists postdating the Variscan

705   orogeny. In contrast, Alpe Sponda sample Sp9712 contains exclusively Alpine monazite

706   that has relic allanite included, indicating an Alpine prograde growth of monazite from

707   allanite.

708   It has been proposed that bulk composition (especially Ca concentration and Ca/Al

709   ratio) plays a key role for the allanite to monazite transition (Wing et al., 2003). This

710   effect may explain allanite stabilisation at lower T in the calcschist than in the quartzite,

711   but this model only holds for the Campolungo samples. CaO contents and Ca/Al ratio do

712   not account for allanite's perpetuation or its replacement by monazite in all samples.

713   The monazite- and allanite- stable areas in Ca/Al space proposed by Wing et al. (2003)

714   do not hold for Central Alps samples studied here (Fig. 9c) and by Janots et al. (2008,

715   2011). This is a strong indication that the reactive bulk composition may play a

716   significant role in accessory mineral stability.

717   The inconsistencies between conclusions reached from the study of natural

718   samples and those from thermodynamic models show that not enough is known about

719   the factors governing allanite and monazite stability to predict the conditions of allanite-

720   in and monazite-in reactions in pelitic to granitic compositions. This reinforces the need

721   for sample-specific petrological considerations (e.g. textural relationships, deformation,

722   effective reactive bulk) in order to link accessory mineral ages with metamorphism.

723   *7.4. Trend of ages across the Lepontine dome*

724   A regional pattern for Barrovian metamorphism develops when the histories of the

725   Croveo-Robiei and Campolungo-Sponda samples are combined with studies from the

726   literature (Fig. 10). Excluding ages from the Southern Steep Belt (SSB), crystallisation

727   ages fall into two groups: older ages (27 to 33 Ma) are interpreted as prograde ages that

728   relate to major, orogen-scale tectonic movements (nappe stacking) and younger ages

729   that can be linked to post-collisional mineral growth close to T_{max}. Prograde allanite ages

730   are in good agreement with the 29-32 Ma Th-Pb allanite ages of Janots et al. (2009) and

731   a greenschist-facies K-Ar amphibole age of 29 ± 3 Ma (Deutsch and Steiger, 1985), both

732   from the Northern Steep Belt (NSB).

733   U-Pb garnet ages of Vance and O'Nions (1992) from the NSB also agree with

734   prograde allanite ages from this study. The garnet investigated by Vance and O'Nions

735   (1992) are re-interpreted as prograde based on the petrological relationships described

736   in that study (i.e. garnet is pre- to synkinematic with the dominant foliation), which are

737   similar to petrological relationships of samples from the current study, in which garnet

738   is a prograde phase.

739   These prograde ages in the northern part of the Lepontine belt coincide with the

740   age of post-HP greenschist-facies overprint dated in Valaisan units in the northeastern

741   Lepontine (Wiederkehr et al., 2009). In the north, progressive regional metamorphism

742   thus reached greenschist-facies conditions within 2-10 Ma after HP metamorphism in

743   the Valaisan units (Wiederkehr et al., 2009), and in Cima Lunga and Adula melange units

744   (Fig. 10, Becker, 1993; Gebauer, 1996; Brouwer et al., 2005; Hermann et al., 2006; Liati

745   et al., 2009). However, the Valaisan units showing HP metamorphism, as well as the

746   mélange units, were exhumed prior to their Barrovian overprint , which postdates

747   nappe emplacement. The correlation of allanite ages with dominant deformation

748   structures suggests that the main deformation event occurred during this time interval,

749   i.e. 32-27 Ma ("stage 2" of Berger et al., 2011); isoclinal refolding of the nappes probably

750   initiated during the later part of this phase. The resolution of ages is not sufficient to

751   evaluate whether there is an E-W age progression in the nappe stacking phase, but a S-N

752   younging of the main deformational phases has been previously established (Berger et

753   al., 2011).

754   The thermal peak followed nappe stacking and maximum ages for this stage are

755   recorded by monazite and a minimum age by Ba0901 allanite rims (Fig. 10). Isograds

756   are clearly discordant to the structures established during the nappe stacking events,

757   providing evidence on a regional scale that thermal metamorphism outlasted this

758   deformation event (Fig. 11a). The regional scale, south-west verging, ductile transverse

759   folding (F4 of Steck et al., 2013) likely occurred close to T_{max}. Combining formation ages

760   that likely record a stage close to maximum T from this study with well-constrained ages

761   from the literature, especially additional monazite ages from further south (Köppel and

762   Grünenfelder, 1975) reveals a consistent trend with metamorphic grade (Fig. 11b). As

763   outlined by the dashed line, younger ages (18-20 Ma) are found in the lower grade rocks

764   (T_{max} = 550-580°C) of the north, and older ages (21-23 Ma) occur in the higher grade

765   rocks (T_{max} = 590-650°C) towards the middle and southern part of the Barrovian

766   sequence.

767   The general trend for crystallisation ages does not apply to rocks of the SSB. Ages

768   for the SSB migmatisation span the entire age range of prograde and T_{max} ages from the

769   subsolidus rocks (Figs. 11 and 12). The spread of ages from 22 to 32 Ma for migmatites

770   has been interpreted to reflect an extended period of upper-amphibolite facies

771   metamorphism with protracted incipient melting during various deformation stages in

772   the SSB (Rubatto et al., 2009; Gregory et al., 2012). Partial melting persisted to 22 Ma,

773   indicating that the rocks resided above the wet solidus (T ≥ 650°C). It should be noted

774   that fission track data (Hurford 1986) show ages around 19 Ma for the partial annealing

775   of zircon in samples from the SSB, reflecting cooling rates >100 °C/Ma for that part of

776   the Lepontine dome.

777   The emerging picture for the age of Barrovian metamorphism for the Lepontine

778   dome is that high temperatures in the southern and central areas lasted to 21-23 Ma,

779   whereas in the north-western part they are slightly younger with ages of 18-20 Ma. The

780   combined age data for main deformations and T_{max} thus suggest that, at least in the

781   central and southern Lepontine, amphibolite-facies conditions were retained for up to

782   10 Ma after nappe stacking, allowing for multiple phases of ductile deformation (Steck et

783   al., 2013). At ~20 Ma, late-orogenic shortening and uplift was accommodated by intense

784   phases of folding, associated with backthrusting in the south, thus producing the two

785   steep belts (SSB and NSB; Berger et al., 2011).

786   Ages of Hurford (1986) and Steck and Hunziker (1994) suggest that rocks of the

787   Croveo-Robiei area had cooled below retention of Rb-Sr in biotite (~ 300 °C) by ~15 Ma

788   and below ~250 °C by ~ 12-15 Ma (zircon fission track). Hurford (1986) also reports a

789   Rb/Sr muscovite age of ~ 18 Ma (T ≈ 500 °C, Hurford, 1986) for samples to the East of

790   Ma9330 (Fig. 10b). Similarly, rapid cooling to below ~ 250 °C by 10-13 Ma is

791   documented for the Campolungo-Sponda area (Hurford, 1986; Janots et al., 2009; Fig.

792   10). Therefore, the Central and Northern part of the Lepontine dome must have

793   experienced very rapid cooling after ~20 Ma.

794   Insight into the cooling history of the eastern samples is provided by Ar-Ar white

795   mica ages from the same samples, as well as one sample from the hanging wall of the

796   Forcola fault (which contain no Alpine allanite). White mica from the hanging wall and

797   footwall of the Forcola fault return Ar-Ar ages of 27-25 Ma and 22-21 Ma respectively

798   (Augenstein, 2013). The younger age in the footwall is interpreted to reflect the onset of

799   grain size reduction and mylonitisation along the Forcola fault (Augenstein, 2013). The

800   Ar-Ar ages also suggest that temperatures were below Ar retention in white mica at 22-

801 21 Ma for the Forcola rocks. K-Ar ages of biotite in this area are around 20 Ma (Steck and

802 Hunziker, 1994) and provide further evidence for rapid cooling. In an east-west

803 traverse, the K-Ar ages of biotite are progressively younger towards the west, suggesting

804 that cooling propagated from east to west (Steck et al., 2013).

805   The accelerated cooling at about 20 Ma, observed throughout the Lepontine dome,

806 did not result in an increased sedimentation rate as would be expected if cooling was a

807 result of increased uplift, erosion and associated exhumation. In contrast, the detailed

808 study of the stratigraphy in the foreland basins surrounding the Lepontine dome reveals

809 that erosion rates decreased by about 50 % at 20 Ma (Schlunegger, 1999). Therefore, the

810 fast cooling around 20 Ma is likely related to exhumation of the Central Alps during a

811 phase of transtensional tectonics (Ciancaleoni and Marquer, 2008). This exhumation

812 might have been accommodated by nearly simultaneous top-to-the-West normal

813 faulting along the Simplon normal fault (Mancktelow, 1992), vertical displacement

814 during back thrusting along the Insubric Line (Schmid et al., 1989) and perhaps normal

815 fault movement on the Forcola line (Ciancaleoni and Marquer, 2008). In any case, the

816 tight age constraints for the transition from low-grade metamorphism to amphibolite-

817 facies conditions across the Central Alps (Figs. 11, 12) provide powerful constraints for

818 the interpretation of this classical orogeny.

819

826   **Table captions**

827   **Table 1:** Sample overview showing the main and accessory mineral assemblage, the

828   minerals used for dating and the results of U-Th-Pb geochronology.

829

830

831   **Figure captions**

832   **Figure 1:** (a) Tectonic overview of the Central Alps and sample locations (after

833   Trommsdorf et al., 2000; Berger et al., 2005; Burri et al., 2005; Maxelon and Mancktelow,

834   2005; Janots et al., 2008). (b) Central Alps map with isotherms for Barrovian

835   metamorphism that overprinted the major tectonic boundaries (Todd and Engi, 1997)

836   and sample numbers. Swiss grid coordinates are in kilometres. For a colour version of

837   the Figure please refer to the online version of the article.

838   **Figure 2:** Photomicrographs of (a) Ma9330 S2 white micas and biotite wrapping

839   garnet (note that S1 and S2 are sub-parallel in these samples); (b) CLB-3 garnet, which

840   preserves internal S1 fabric, rotated and wrapped by S2 white mica and biotite; (c)

841   Monazite crystal in CLM-6 that is aligned along the main foliation defined by micas and

842   graphite; (d) Ma0901 allanite aligned along the S1/S2 fabric; (e) Sp9712 large staurolite

843   crystals surrounding kyanite that is partially replaced by white mica; (f) LEP0980

844   allanite aligned along the dominant fabric (S1). A detailed petrographic description of

845   the samples is given in the electronic Appendix.

846   **Figure 3:** (a) Backscatter electron (BSE) image and (b) compositional map of

847   Ba0901 allanite showing an allanite rich-core (C), epidote mantle (M) and allanite rim

848   (R). BSE images of (c) CLB-3 allanite; (d) Ma0901 allanite, arrows point to rutile

849   inclusions in the allanite core; (e) Sp9712 monazite satellites of a relic allanite; (f)

850   LEP0979 allanite rims on epidote-rich cores; (g) LEP0807 allanite; (h) LEP0980 mosaic

851   allanite and regularly zoned allanite (P = pre-Alpine relics).

852   **Figure 4:** Garnet composition (data from Table A2). Ma9330 garnet: (a) MgO,

853   MnO and Lu profile across the garnet; O. Rim = outer rim of the garnet; (b) chondrite-

854   normalised REE, the systematic depletion in HREE from core (C) to rim (R) is indicated

855   by the arrow; outer rim garnet is mildly enriched in MREE. (c) MgO, MnO and Lu profile

856   across CLB-3 garnet, showing a sharp increase in Lu from Grt I rim to Grt II; (d)

857   chondrite-normalised REE patterns of CLB-3 garnet. (e) MgO, MnO and CaO profile

858   across CLM-6 garnet. The co-rim boundaries (vertical dashed lines) are based on the

859   element map shown in (f). (f) element distribution map of Ca in garnet from CLM-6. Note

860   the Alpine rim cutting across the regular zoning in the pre-Alpine core. (g) LA ICPMS

861   profile across Sp9712 Grt I and II; location of the profile is given in the Ca map (inset). I_C,

862   I_R = Grt I core and rim respectively, II_C = Grt II core etc. (f) Sp9712 chondrite-normalised

863   REE patterns of garnets I-III. Inset shows Ca map of Grt III. The decrease in HREE from

864   core to rim is indicated.

865   **Figure 5:** Chondrite-normalised REE composition of accessory allanite (a-f) and

866   monazite (g-h) compared with whole rock (WR). REE of allanite cores, mantles and rims

867   of (a) Ba0901, (b) CLB-3 and (c) Ma0901. (d) A comparison of allanite growth zones in

868   terms of REE and Th/U in metasediments. REE of allanite in Forcola orthogneiss

869   samples (e) LEP0807 and LEP0979 and (f) LEP0980. (g) REE of monazite in Croveo

870   schist (Ma9330) and Alpe Sponda fels (Sp9712). (h) REE compositional variation in

871   Alpine monazite. The effect of feldspar and garnet coexistence is indicated with arrows.

872   **Figure 6:** Summary of geochronology results grouped by area (Western, Central

873   and Eastern Lepontine) and by phase dated. Monazite and allanite ages are formation

874   ages, whereas rutile dates are more difficult to interpret, see text for discussion.

875   **Figure 7:** Monazite ages. (a) Concordia plot for monazite analyses in samples CLM-

876   5 and CLM-6 (a), in sample Ma9330 (b) and in sample Sp9712. Average $^{206}Pb/^{238}U$ ages

877   are given in (a), concordia ages are given in (b) and (c) and are represented by the blue

878   ellipses. The unfilled ellipses in (a) and (c) are not used for age calculation. A

879   representative BSE image of dated monazite is shown on the right of plot (a) and (c).

880   **Figure 8:** Allanite Th-Pb isochrons for (a) Ba0901 allanite cores; (b) Ba0901

881   allanite rims; (c) CLB-3 allanite cores; (d) Ma0901 allanite cores; (e) LEP0980 mosaic

882   allanite cores; (f) LEP0807 allanite (cores and rims). The model common Pb

883   composition (Stacey and Kramers, 1975) is indicated on the y-axis and always close to

884   the intercept defined by the isochron. Error ellipses are 2σ; errors quoted on isochron

885   ages are 95 % confidence.

886   **Figure 9:** Allanite- and monazite-bearing samples of Alpine age compared to

887   modelled stability fields. (a) Samples plotted in terms of temperature and bulk-rock CaO

888   with stability fields calculated by Spear (2010). Numbers next to symbols are wt %

889   Al_2O_3. Reaction lines of Spear (2010) were calculated for 5 kbar pressure; the reaction is

890   predicted to occur at higher temperature with increasing bulk CaO, and the slope is

891   dependent on bulk Al_2O_3. (b) Samples plotted in the allanite-monazite P-T stability field

892   of Spear (2010). Numbers indicate the sample whole rock CaO concentration (wt %);

893   more than one number next to a symbol indicates more than one sample from the same

894   locality. Two reaction curves are plotted: one for low CaO and one for moderate Ca

895   concentration. (c) Al/Al$_{Shaw}$ vs. Ca/Ca$_{Shaw}$, where Al$_{Shaw}$ and Ca$_{Shaw}$ are concentrations for

896   an "average pelite" (Shaw, 1956). The discrimination line of Wing et al. (2003) shows

897   the expected stability of allanite and monazite for metapelites in andalusite, kyanite and

898   sillimanite zones. None of the diagrams predicts correctly the allanite or monazite

899   presence in the studied samples.

900   **Figure 10:** Summary of Alpine metamorphism in the Central Alps. (a) An overview

901   of interpretation of metamorphic ages in the Lepontine Alps; (b) Temperature-time

902   paths for samples studied as well as for the Southern Steep Belt (SSB), Northern Steep

903   Belt (NSB) and the Urseren zone, taken from the literature as indicated. (1) Becker,

904   1993; (2) Gebauer, 1996; (3) Brouwer et al., 2005; (4) Wiederkehr et al., 2009; (5)

905   Deutsch and Steiger, 1985; (6) Janots et al., 2009; (7) Vance and O'Nions, 1992; (8)

906   Janots and Rubatto, 2014; (9) Köppel and Grünenfelder, 1975; (10) Hurford, 1986; (11)

907   Liati et al., 2000; (12) von Blanckenburg, 1992; (13) Oberli et al., 2004; (14) Gregory et

908   al., 2012; (15) Rubatto et al., 2009; (16) Augenstein, 2013.

909   **Figure 11:** Trend of ages across the Lepontine dome. (a) Map of isotherms with

910   locations of samples included in (b). Note that the study site of Janots and Rubatto

911   (2014) is north of the map area. (b) Metamorphic grade (T_{max}) vs. allanite, monazite and

912   zircon crystallisation age from this and selected studies from the literature as indicated.

913   Legend is the same for both (a) and (b).

914

915

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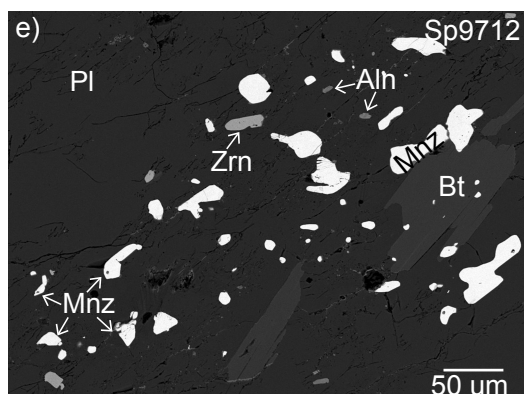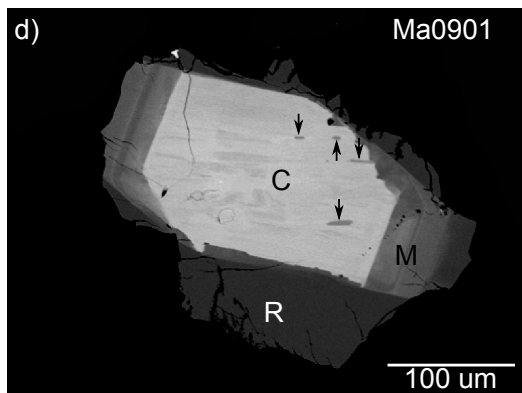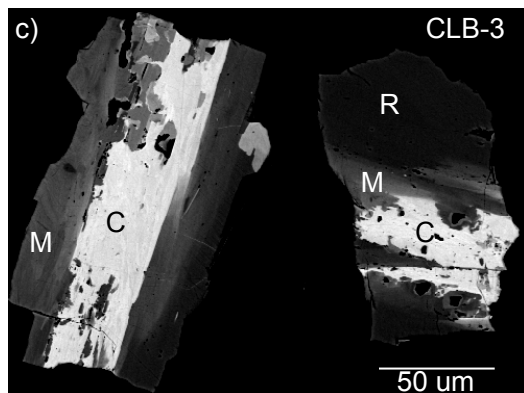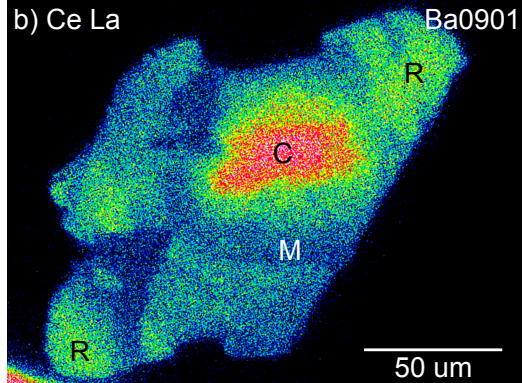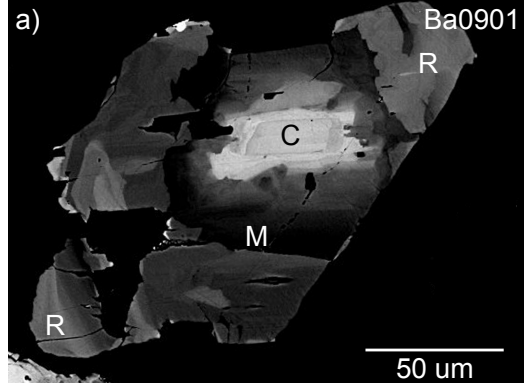1153   the Alpine orogenic wedge. Swiss Journal of Geoscience, 101, S127-155.

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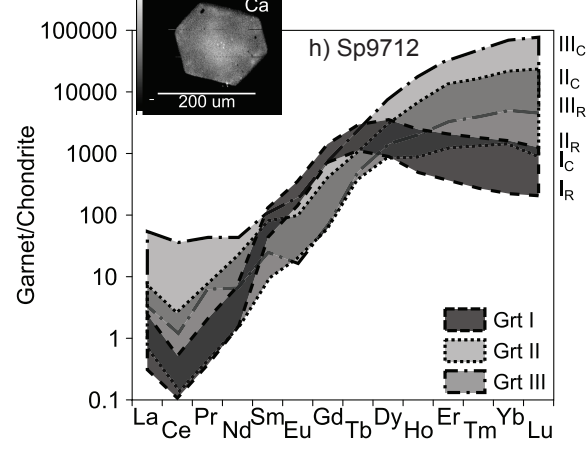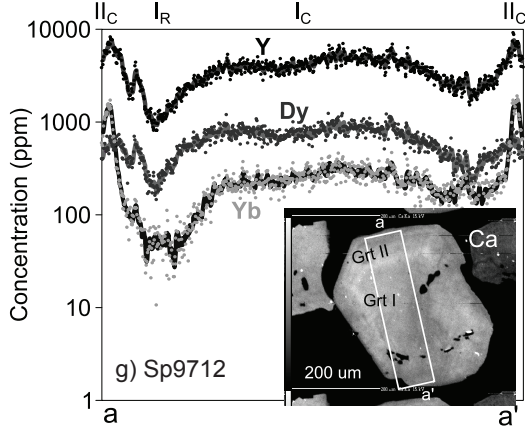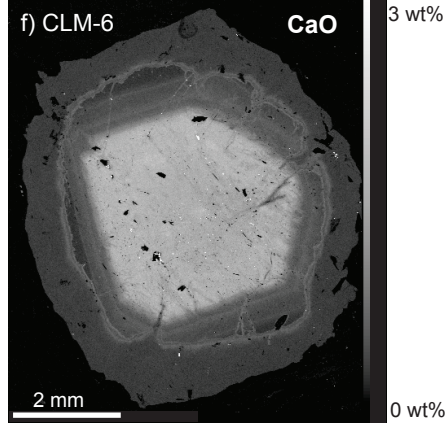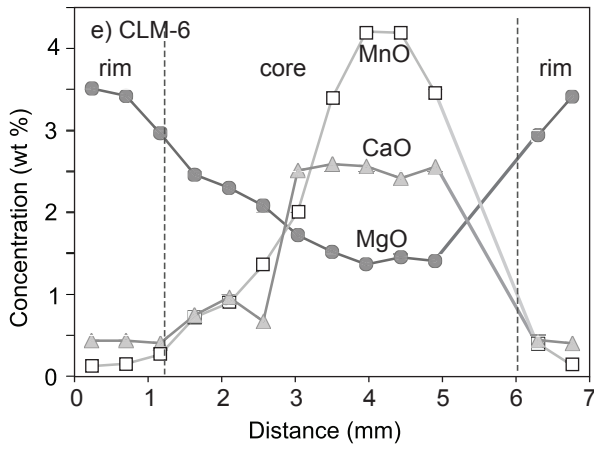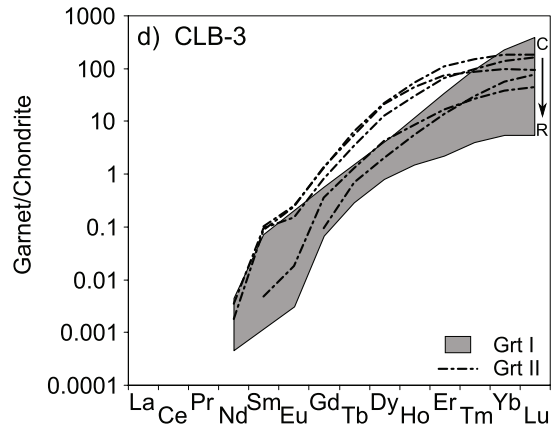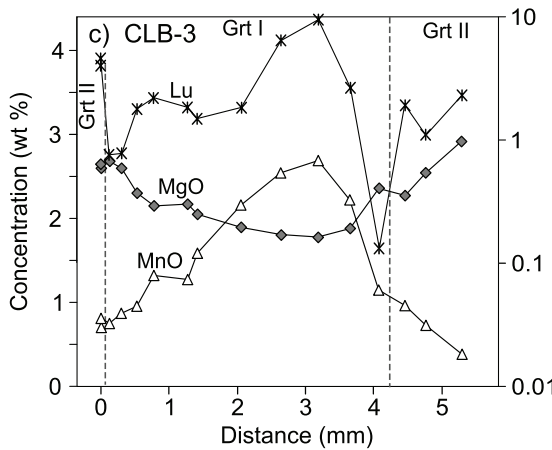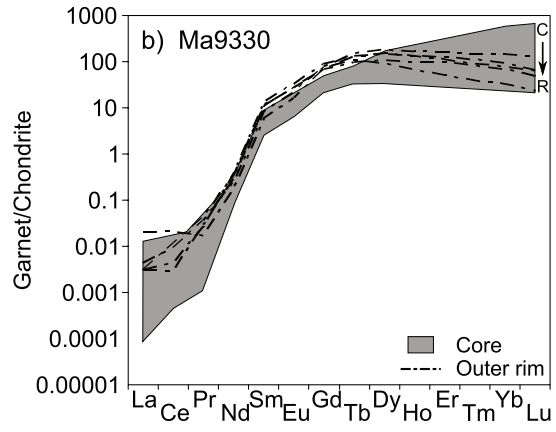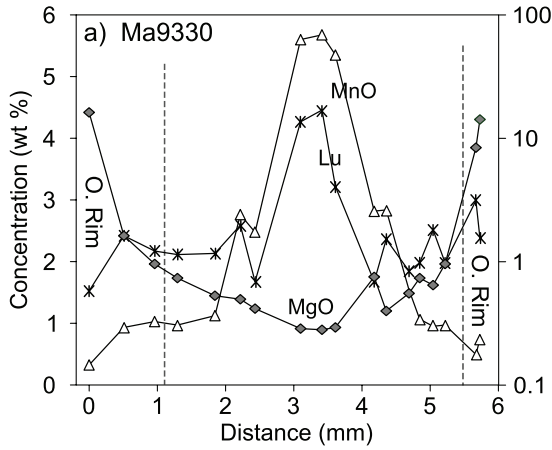1157   Wiederkehr, M., Bousquet, R., Ziemann, M. A., Berger, A., Schmid, S., 2011. 3-D
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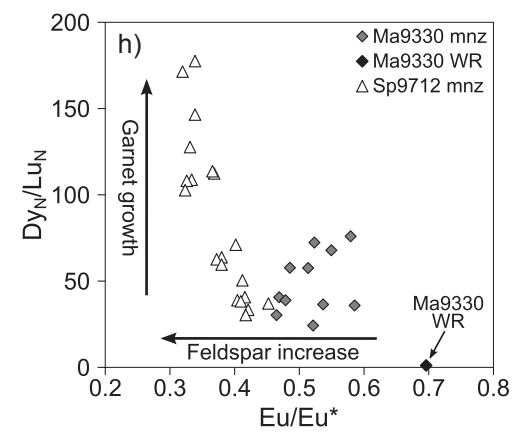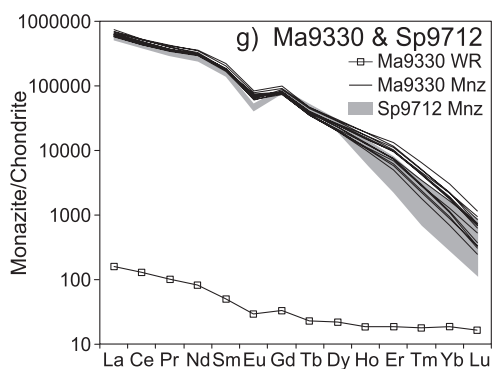1161   Wing, B.A., Ferry, J.M., Harrison, T.M., 2003. Prograde destruction and formation of
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a)

Central Alps:
- Polycyclic basement
- Valais units
- Brianconnais sedimentary cover
- European sedimentary cover
- Tectonic melange
- Tertiary intrusives

Major structures:
FF Forcola Fault

◇ Monazite-bearing samples

△ Allanite-bearing samples

b)

·— Isotherms:
25° contour interval

a) Ma9330 — St, S2, Ky, Wm + Bt, Grt, Qtz

b) CLB-3 — Wm, S2, S1, Wm + Bt, Grt, Qtz, S2, Grt, S2, Bt

c) CLM-6 — Wm + Bt, Mnz

d) Ma0901 — Tur, Grt, Wm, Aln, Ep, S1/S2, Qtz

e) Sp9712 — St, Wm, Wm, Ky, Wm, St, Bt, St

f) LEP0980 — Qtz + Fsp, Chl, Aln, S1, Bt

a) Ma9330
O. Rim
MnO
Lu
MgO
O. Rim

b) Ma9330
Core
Outer rim

c) CLB-3   Grt I   Grt II
Grt II
Lu
MgO
MnO

d) CLB-3
C
R
Grt I
Grt II

e) CLM-6
rim   core   rim
MnO
CaO
MgO

f) CLM-6   CaO
3 wt%
2 mm
0 wt%

g) Sp9712
II_C I_R I_C II_C
Y
Dy
Yb
a
a'
Grt II
Grt I
Ca
200 um

h) Sp9712
Ca
200 um
III_C
II_C
III_R
II_R
I_C
I_R
Grt I
Grt II
Grt III

Western Lepontine

Allanite — Ba0901 rims ... Ba0901 cores

Monazite — Ma9330

Rutile — Ba0903A

Central Lepontine

Allanite — Ma0901 ... CLB-3

Monazite — Sp9712

Rutile — CLB-4 ... Sp9712

Eastern Lepontine

Allanite — LEP0979 / LEP0980 / LEP0807

Age (Ma)

a)
CLM5: 331 ± 4 Ma, N 13, MSWD 1.6
CLM6: 326 ± 4 Ma, N 9, MSWD 0.25
error ellipses are 2sigma
Pb disturbance

b)
Ma9330 concordia age 21.7 ± 0.4 Ma
N 18
error ellipses are 2sigma

c)
Sp9712 concordia age 22.0 ± 0.4 Ma
N 18
error ellipses are 2sigma

a) Ba0901 cores

error ellipses are 2σ

Age: 26.8 ± 2.3 Ma
Initial ^{208}Pb/^{206}Pb = 2.174 ±0.048
MSWD = 2.0, n = 9

^{208}Pb/^{206}Pb$_c$
^{232}Th/^{206}Pb$_c$
SK

b) Ba0901 rims

error ellipses are 2σ

Age: 19.7 ± 1.3 Ma
Initial ^{208}Pb/^{206}Pb = 2.061 ±0.060
MSWD = 0.77, n = 11

^{208}Pb/^{206}Pb$_c$
^{232}Th/^{206}Pb$_c$
SK

c) CLB-3 cores

error ellipses are 2σ

Age: 31.7 ± 1.1 Ma
Initial ^{208}Pb/^{206}Pb = 2.083 ±0.015
MSWD = 1.5, n = 15

^{208}Pb/^{206}Pb$_c$
^{232}Th/^{206}Pb$_c$
SK

d) Ma0901 cores

error ellipses are 2σ

Age: 28.5 ± 1.3 Ma
Initial ^{208}Pb/^{206}Pb = 2.084 ±0.014
MSWD = 0.60, n = 27

^{208}Pb/^{206}Pb$_c$
^{232}Th/^{206}Pb$_c$
SK

e) LEP0980 mosaic cores

error ellipses are 2σ

Age: 31.3 ± 1.1 Ma
Initial ^{208}Pb/^{206}Pb = 2.086 ±0.014
MSWD = 1.1, n = 16

^{208}Pb/^{206}Pb$_c$
^{232}Th/^{206}Pb$_c$
SK

f) LEP0807

error ellipses are 2σ

Age: 27.4 ± 0.6 Ma
Initial ^{208}Pb/^{206}Pb = 2.115 ±0.044
MSWD = 1.9, n = 17

^{208}Pb/^{206}Pb$_c$
^{232}Th/^{206}Pb$_c$

a)

Allanite

Monazite

24.86% Al2O3

12.43% Al2O3

16.95% Al2O3

△19.10
△16.27

△15.26

12.23
△
13.16△

◆18.70

△6.59

b)

+ Qtz
+ H₂O
+ Al₂SiO₅
+ Garnet
+ Apatite

Allanite

Monazite

0.74△   ◆0.94

△0.13,
2.54

0.94,△
1.72,
2.38

1.08 wt% CaO

Aln & Mnz

2.17 wt% CaO

c)

Allanite stable

Monazite stable

Wing et al. 2003

◆Ma9330   △ CLB-3

△ LEP0980

△LEP0979

△Ba0901
△LEP0807

△Ma0901

△ Allanite-bearing sample
◆ Monazite-bearing sample

a)

Elogite facies (Adula, Cima Lunga)
△ This study
(1,2,3)

Main deformations
(4,5,6,7)

Temparature peak
(6,8,9)

Cooling below 250°C (central and N Lepontine)
(6,10)

Intrusions
Novate (9,11)   Bergell (12,13)

Partial melting, SSB
(14,15)

Age (Ma)

b)

Croveo
Robiei
Campolungo
Sponda
Forcola
SSB(14,15)
NSB(6)
Urseren(8)

HP stage (1,2,3)

Zrn U-Pb (15)
Aln Th-Pb (14)

Wm Ar-Ar (16)

Bt K-Ar, Rb-Sr (10)

Zrn FT (4,10), Mnz U-Pb (8)

Ap FT (6,10)

T (°C)

Age (Ma)

a)

Elogite facies (Adula, Cima Lunga)
Main deformations
Temparature peak
Cooling below 250°C (central and N Lepontine)
Intrusions
Partial melting, SSB

△ This study

(1,2,3)
(4,5,6,7)
(6,8,9)
(6,10)
Novate (9,11) Bergell (12,13)
(14,15)

Age (Ma)

b)

Croveo
Robiei
Campolungo
Sponda
Forcola
SSB[14,15]
NSB[6]
Urseren[8]

HP stage (1,2,3)

Zrn U-Pb (15)
Aln Th-Pb (14)

Wm Ar-Ar (16)

Bt K-Ar, Rb-Sr (10)

Zrn FT (4,10), Mnz U-Pb (8)

Ap FT (6,10)

T (°C)

Age (Ma)

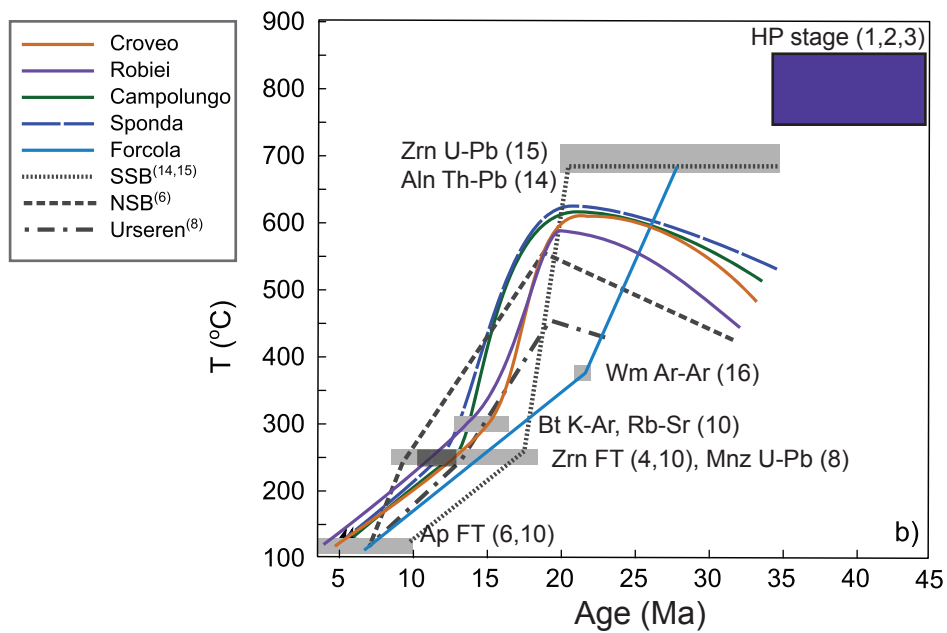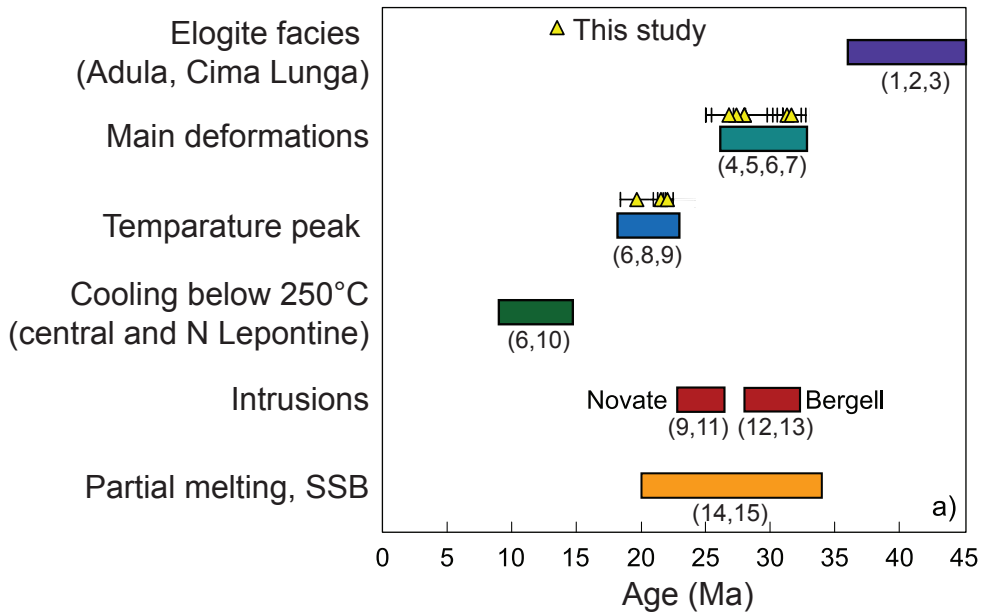**Table 1:** Sample overview showing the main and accessory mineral assemblage, the minerals used for dating and the results of U-Th-Pb geochronology.

Sample	Swiss Grid coordinates X	Y	Location	Rock type	Mineral assemblage Main	Accessory	Mineral dated	Age (Ma)
Ma9330[1]	666.51	124.88	Croveo, (N of Domodossola)	Metapelite	Qtz Wm Bt Pl Grt St Ky	Mnz Ap Rt Ilm Zrn	Mnz	21.7 ± 0.4
Ba0903A	682.20	143.37	Robiei	Metapelite "Bündnerschiefer"	Qtz, Wm Bt Pl Grt Cal	Aln Rt Ilm Zrn	Rt	(21–27)[3]
Ba0901	683.23	142.70	Robiei	Gneiss	Qtz Wm Bt Kfs Pl	Aln Ap Zrn Rt	Aln cores	26.8 ± 1.3
							Aln rims	19.7 ± 1.3
CLB-3	697.37	147.60	Campolungo	Metapelite "Bündnerschiefer"	Qtz Wm Bt Pl Grt	Aln Rt Zrn	Aln	31.7 ± 1.1
CLB-4	697.37	147.60	Campolungo	Metapelite "Bündnerschiefer"	Qtz Wm Bt Pl Grt Ky St	Aln Rt Zrn	Rt	(19–25)[3]
CLM-5	698.04	146.94	Campolungo	Metasediment, basement	Qtz Wm Bt Pl Grt St Ky	Zrn Mnz Tur	Mnz	332 ± 4
CLM-6	698.82	146.66	Campolungo	Metasediment, basement	Qtz Wm Bt Pl Grt St	Zrn Mnz	Mnz	326 ± 4
Ma0901	698.29	147.23	Campolungo	Metaquartzite	Qtz Wm Bt Pl Grt	Ap Aln Rt Zrn	Aln	28.5 ± 1.3
Sp9712	702.78	142.20	Alpe Sponda	Metasediment, basement	Qtz Wm Bt Pl Grt St Ky	Mnz Rt Zrn	Mnz	22.0 ± 0.4
							Rt	(57–33)[3]
LEP0979[2]	743.95	130.70	Forcola	Orthogneiss	Qtz Wm Bt Kfs Pl	Aln Zrn	Aln	25–31
LEP0980[2]	744.05	131.24	Forcola	Orthogneiss	Qtz Wm Bt Chl Kfs Pl	Aln Zrn	Mosaic Aln	31.3 ± 1.1
							Regular Aln	27–31
LEP0807[2]	744.18	131.38	Forcola	Orthogneiss	Qtz Wm Bt Chl Kfs Pl	Aln Zrn	Aln	27.4 ± 0.6

[1] Ma9330 is the sample used in Todd & Engi, 1997

[2] LEP0807, -0979 and -0980 were donated by C Augenstein

[3] These are not formation ages, but dates that may include inheritance and Pb loss after formation, see text for discussion.

Appendix

1. Sample description

<u>Western Samples: Croveo (Ma9330) and Robiei (Ba0901, BA0903A)</u>

Sample Ma9330 is the highest-grade rock of the western samples and reached T_{max} of ~ 620 °C (617± 11°C for this sample in Todd and Engi, 1997). The rock is characterised by inclusion-rich garnet porphyroblasts, rotated in the foliation (Fig. 2a), and rounded staurolite and kyanite (see also sample description in Todd and Engi, 1997). Inclusions in garnet are mainly quartz with minor ilmenite and relic allanite. Allanite is not present in the matrix; monazite, which displays no zoning in BSE images, is a matrix mineral and has not been observed as inclusion in garnet. The bulk rock composition is similar to that of the Campolungo calcschist (Bündnerschiefer CLB-3), except that it has a lower CaO concentration and Ca/Al ratio (Table A1).

Three foliations are present in Ma9330. In schist Ma9330 S1 is a relic foliation defined by the alignment of kyanite and staurolite, overprinted by a sub-parallel S2 fabric defined by mica that wraps around kyanite, staurolite and garnet (Fig. 2a). A texturally later weak foliation (S3) is defined by sparse biotite that tends to be smaller than S2 biotite.

Robiei samples Ba0901 and Ba0903A are from an area that reached significantly lower T_{max} than Ma9330 during Barrovian metamorphism (T_{max} ~ 575 °C according to the regional T distribution of Todd and Engi, 1997). Calcschist Ba0903A contains white mica, biotite, plagioclase and large poikiloblastic garnets (similar to those found in Ma9330), but lacks both staurolite and kyanite. Minor calcite occurs in low-strain bands comprised

predominantly of quartz. Ba0901 is a fine-grained gneiss containing mm-sized porphyroblasts of K-feldspar and quartz, wrapped in a matrix of feldspars, quartz, biotite and muscovite. The bulk composition of Ba0901 gneiss closely resembles the orthogneiss samples in terms of Al and Na and is low in Ca and Ca/Al (CaO = 0.7 wt%, Ca/Al = 0.08; Table A1). Ba0901 allanite is characterised by cores with high BSE emission that are surrounded by mantle zones, which are overgrown by allanite rims (Fig. 3a-b). The mantle domains change from allanitic compositions near the core, to more epidote compositions towards the rim. The allanite core and epidote mantle are partially replaced by a second generation of allanite at the rim; allanite rims form a sharp contact with the mantle. Both core and rim allanite have inclusions of muscovite and biotite.

In Ba0903A, S1 is a relic foliation that is overprinted by sub-parallel S2. S1 is defined by the alignment of mica and allanite-epidote that is preserved in garnet; S2 is distinguished from the relic foliation as it wraps around garnet. S3 a weak foliation that is defined by a few biotite grains and crosscuts S1 and S2 at a high angle. Ba0901 displays only one strong foliation (likely to be S1/S2), which is defined by the alignment of muscovite and biotite, and that also contains allanite. The foliation is overgrown by a second generation of larger grains of biotite and white mica that have no preferred orientation.

<u>Central samples: Campolungo (CLB-3, CLB-4, Ma0901, CLM-5 and CLM-6) and Alpe Sponda (Sp9712)</u>

Calcschists CLB-3 and CLB-4, and impure quartzite Ma0901 are from the Campolungo area, southeast of Airolo (Fig. 1). CLB-3 represents a pelitic layer within the Bündnerschiefer. It has the highest CaO concentration of the samples

analysed for bulk composition (CaO = 2.5 wt %) and a high Ca/Al ratio of 0.18 (Table A1). Typical of Bündnerschiefer, CLB-3 hosts mm-to-cm sized garnet porphyroblasts in a fine-grained grey-blue matrix that is predominantly biotite, white mica, plagioclase and quartz (Fig. 2b). Accessory allanite occurs as cores in large (> 200 μm) epidote grains that have three distinct zones: small cores (< 100 μm) with high BSE emission, a mantle zone (< 50 μm or absent) of lower emission, and an epidote rim (Fig. 3c). In sample CLB-3, S1 is a relic foliation defined by the alignment of mica and allanite preserved in garnet (Fig. 2b). S2 is the dominant foliation and it is defined by biotite and white mica that wrap garnet and allanite-epidote grains. S2 is folded and axial-planar biotite and white mica form a third foliation (S3). CLB-4 contains staurolite and kyanite in addition to garnet, white mica, biotite, plagioclase, and accessory allanite, rutile and zircon. Textural relationships and foliations are similar to CLB-3. In CLB-4, staurolite and plagioclase both overgrow folded white mica and biotite.

Ma0901 is a mica-rich impure quartzite (SiO_2 = 88 %) with low CaO content (0.13 wt. %) and Ca/Al = 0.03 (Table A1). This sample was collected between the prominent dolomite marble bands representing the Triassic sediments and the Simano basement (Fig. 1a). The protolith of Ma0901 is most likely a Permian sandstone. Bands of muscovite host tourmaline, rutile and allanite-epidote (Fig. 2d). Garnets are small, rare and occur exclusively in low-strain bands of quartz; no relic foliation is preserved in the small garnets. Similar to CLB-3 calcschist allanites, Ma0901 allanite grains display three distinct growth zones (Fig. 3d). Large cores (up to ~ 300 μm) are host to rutile, muscovite and biotite inclusions. Mantle zones have intermediate BSE emission and often show oscillatory zoning; epidote rims have low BSE emission. There is

one foliation preserved in Ma0901, which is defined by the alignment of muscovite and allanite (Fig. 2d). This foliation is likely to be S1/S2 composite (as S1 and S2 are sub-parallel in the other samples) and it is overgrown by late-stage biotite, chlorite and plagioclase, none of which tend to have a preferred orientation.

CLM-5 and 6 are two staurolite-garnet schists collected a few hundred meters from each other within the Simano nappe. CLM-5 is close to the locality "Fontane" located within the Prevat synform. CLM-6 comes from the Southern end of Laghetto di Campolungo. The metasedimentary rocks in this area have been attributed to late Paleozoic sediments (Permo-Carboniferous) that display chemical characteristics compatible with an active continental margin setting (Gieré et al. 2011). The two samples have a typical amphibolite-facies assemblage. At the cm-dm scale, the samples contain numerous deformed quartz layers alternated by darker layers with a fine-grained crenulated foliation that is defined by muscovite, biotite, quartz, minor plagioclase, ilmenite and graphite. Porphyroblastic garnet is partly wrapped by the main foliation (likely S2), which is folded. Staurolite and kyanite (CLM-5) crystals overgrow the foliation preserving remnants of foliation defined by graphite. CLM-5 also contains abundant tourmaline. Monazite grains are aligned along the main foliation and also contain inclusions of graphite (Fig. 2c).

Sample Sp9712 is from the Alpe Sponda area, a key locality for Alpine kyanite. The sample is from the base of Monte Forno, situated within the Campo Tencia polymetamorphic basement that is considered part of the Simano nappe (Steck et al. 2013). The locality also exposes ortho-amphibolites and calc-silicate

nodules, which are typical of basement units in the gneiss nappes. Sp9712 is a coarse-grained granofels that has two compositional layers: one predominantly kyanite and staurolite, with muscovite, paragonite, and minor biotite and plagioclase; the other is almost exclusively biotite and plagioclase. The staurolite-kyanite layer is characterised by millimetre-to-centimetre scale staurolite and kyanite. Two generations of kyanite are apparent and the older kyanite grains are partly replaced by white mica (Fig. 2e); the second kyanite generation displays sharp grain edges and shows no signs of replacement. Sp9712 garnet is relatively small (< 500 μm), sparse, and occurs only in plagioclase within the biotite-rich compositional layer. Garnet contains inclusions of relic allanite but not monazite. Unlike the samples from Campolungo, Sp9712 has no dominant foliation in thin section or hand-specimen scale: large staurolite and kyanite are randomly orientated, as are the white mica and biotite grains in both compositional domains.

Accessory rutile and monazite occur in both compositional layers in Sp9712. Rutile occurs in the matrix and as inclusions in staurolite and garnet. Monazite often occurs in groups of more than five grains, sometimes as satellites surrounding a small (~ 10 μm) relic allanite grain (Fig. 3e). Monazite can also contain inclusions of relic allanite. Monazite grains are subtly heterogeneous in BSE images and display either patchy or core-rim zoning.

Eastern samples: Forcola orthogneiss (LEP0979, LEP0980, LEP0807)

The eastern-most samples are orthogneisses from the Adula nappe in the footwall of the Forcola fault (Fig. 1). Samples LEP0979 and LEP0807 are predominantly quartz, K-feldspar, and white mica with minor plagioclase and

biotite, chlorite, and accessory allanite and zircon. LEP0980 differs only in that biotite predominates and white mica is a minor component. In the three samples the major foliation (S1) is defined by the alignment of white mica (LEP0979 and LEP0807) or biotite (LEP0980, Fig. 2f), which are overgrown by larger micas belonging to a later metamorphic stage. In all three samples, allanite is aligned along the major foliation (S1). Neither garnet nor rutile is present in any of the orthogneiss samples. Forcola orthogneiss has CaO concentrations that vary from 0.9–2.4 wt % (Table A1). Ca/Al ratios are at the higher end of the analysed values (0.1–0.2).

LEP0979 and LEP0807 allanite occurs as overgrowths with high BSE emission on patchy or oscillatory-zoned epidote grains (Fig. 3f). LEP0807 allanite can also form large cores in patchy grains (Fig. 3g). Sample LEP0980 contains allanite with a variety of textures (Fig. 3h). Pre-Alpine igneous allanites (~250 Ma, see below) are weakly zoned in BSE images and can have small, presumably metamorphic overgrowths. Alpine allanites have vastly different internal zoning and are divided into two groups: mosaic and regularly zoned allanites. Regularly zoned allanite can occur as single grains, locally with weak oscillatory zoning or as overgrowths on epidote grains. Unique "mosaic" grains are aggregates of several small allanite sub-grains. Mosaic allanites are further subdivided into early (mosaic core) and late (mosaic rim) domains, according to the internal zoning and overprinting relationships within the subgrains (C and R in Figure 3h).

2. Methods

Bulk rock analysis

Bulk rock analysis was conducted at the Central Analytical Facilities, Stellenbosch University, South Africa. Samples were milled in an agate mill before being mixed with CLAISSE flux (1 g sample to 10 g flux) and fused into a lithium borate disc. Si, Ti, Al, Fe, Ca, Mg, Mn, Na, K and P were measured by XRF of fused discs using an Axios (PANanytical) with a 2.4 kWatt Rh tube. Trace elements were analysed by LA ICPMS (method outlined below).

Scanning Electron Microscope (SEM) and electron microprobe

Back scattered electron (BSE) images of minerals in thin section and polished grain mounts were made using the JEOL 6400 SEM (15 kV, WD 39 mm), the Cambridge 360 (20kV, 3nA, WD 15 mm) at the Centre for Advanced Microscopy at the Australian National University (ANU), and the JEOL JSM 6610A (15 kV, WD 10-15 mm) at the Research School of Earth Sciences (RSES), ANU. The EDS-detector on the JEOL 6400 was also used for mineral identification and for quantitative x-ray analysis of major phases.

Electron microprobe spot analyses of allanite and element maps of garnet were made using the Cameca SX 100 at the ANU. Accelerating voltage was 15 kV with a current of 100 nA. Elements analysed were Si (K-α), Ti (K-α), Al (K-α), Fe (K-α), Mn (K-α), Mg (K-α), Ca (K-α), Y (L-α), La (L-α), Ce (L-α), Pr (L-β), Nd (L-β), Sm (L-β), Th (M-α) and U (M-β). The background positions were carefully selected after line-scans on trace element-rich allanites in order to avoid interferences. Daibosatsu allanite (Hoshino et al. 2005) and Fukudayama allanite

(Hoshino et al. 2006) were used as secondary standard to check within-session variability. Values for Daibosatsu allanite were within 1 wt % of published values for all elements analysed, whereas Fukudayama allanite analyses were within 1 wt % for elements measured except for Si, Fe and Th. Si and Th were within 1.5 wt % and Fe was within 3 wt % of published values (Hoshino et al. 2006). The same beam set up conditions were used for making element maps of garnet at ANU. The dwell time on each spot was 100 ms with a step interval of 1 μm.

LA-ICP-MS trace element analysis

Trace element analysis was made on polished epoxy grain mounts, thin sections and bulk rock glasses using an Agilent 7500s and 7700 quadrupole inductively coupled plasma mass spectrometer (ICP-MS) in peak-jumping mode, attached to an ArF excimer laser (193 nm wavelength, at 5 Hz) system at RSES. Samples were loaded into a purpose-built HelEx ablation cell and ablation was carried out in a He atmosphere (Eggins et al. 1998). An analysis consisted of 20-30 seconds of background gas analysis, followed by 30-45 seconds of ablation. Contamination by inclusions, drill through or alteration was determined by monitoring several pertinent elements during the time-resolved analysis and only the relevant part of the analysis was included in the integration.

Analyses of bulk rock glasses were made using a 108 or 178 μm spot. The analysis spot diameter was 22 μm for allanite and monazite, 47 or 81 μm for garnet, 37 or 47 μm for rutile depending on the size of the grains.

Traverses of Sp9712 garnets were made by scanning a slit ~10 μm long across the sample at a rate of ~ 1 μm/second (Spandler et al. 2007). Where possible, the slit was aligned parallel to internal zoning of major elements in the

garnet, as revealed by X-ray maps. Synthetic glass standards NIST 610 and 612 were used, with BCR, a natural glass, as a secondary standard. Reference values of Pearce et al. (1997) were used for NIST 612, and from Spandler et al. (2011) for NIST 610. The standard glass was analysed routinely every 10 unknowns. The secondary standard (BCR) was analysed every ~ 20 unknowns and was within 5 % of reference values for elements Ti, V, Mn, Sr, Nb, Ba, La, Pr, Nd, Sm, Eu, Ta, Th, U; within 10% for elements P, Sc, Cr, Zr, Cs, Ce, Gd, Dy, Ho, Er, Yb, Lu, Hf, Pb; and within 15% for Y, Tb and Tm. SiO_2, as determined by electron microprobe, was used as internal standard for allanite, epidote and garnet; stoichiometric Ce and Ti were internal standards for monazite and rutile, respectively (219700 ppm Ce; 599000 ppm Ti). SiO_2 ascertained from XRF analyses was used as the internal standard for bulk rock glasses. Data reduction used the in-house excel spreadsheets written by C Allen, and Iolite (Paton et al. 2011).

SHRIMP U-Th-Pb analysis

Prior to isotope analysis, allanite and monazite were imaged by BSE using the Cambridge 360 and the JEOL JSM 6610A and conditions described above. Allanite was analysed for U, Th and Pb isotopes using SHRIMP RG at RSES. Polished epoxy mounts of standards and unknown grains were analysed by a primary O$^-$ ion beam, focussed to a ~ 20 μm spot. Analysis of allanite is similar to zircon (Williams 1998) albeit with the addition of mass 232 (Th) to the run table and the use of the mass peak at 198.9 amu ($^{139}La^{28}Si^{16}O$) as the reference peak (Gregory et al. 2007). An analysis consisted of six scans through each of the masses. The standardisation procedure to correct for instrumental mass fractionation followed Gregory et al. (2007) and was based on the Th/ThO

versus $^{208}Pb/^{232}Th$ correlation instead of the U/UO versus $^{206}Pb/^{238}U$ calibration scheme that is commonly used for zircon and monazite.

Two allanite standards were used in each of the analytical sessions. The primary standard TARA (417 Ma; Gregory et al. 2007) was analysed every three unknowns. Calibration errors in each session were between 1.5 and 1.8 % (1σ). The secondary standard BONA (ID-TIMS Th-Pb age: 30.1 ± 0.3 Ma; von Blanckenburg 1992) was used to gauge accuracy. With the exception of three anomalous analyses on domains that displayed subtle patchy zoning, BONA returned a weighted average age of 29.77 ± 0.44 Ma (95 % conf., MSWD = 1.5, n = 14/17).

A significant proportion (up to 90%) of the Pb present in metamorphic allanite may be from initial incorporation, and not from the *in situ* decay of U and Th and thus correction for initial Pb becomes extremely important to obtain meaningful ages. Initial Pb (Pb_c) is used here to mean the Pb component initially incorporated into the crystal, which includes common Pb as well as radiogenic Pb inherited from precursor phases. The Th-isochron method is used as it has been demonstrated to give reliable ages that are largely independent of the Pb_c composition, (Gregory et al. 2007; Janots and Rubatto 2014). The initial estimation of $^{206}Pb_c$ and construction of isochrons follows the method outlined by Gregory et al. (2007) and Janots and Rubatto (2014). ^{207}Pb-corrected single spot dates are presented only where the data do not form a well-constrained isochron, but their geological significance is considered unreliable. The ^{207}Pb-correction used assumes the model common Pb composition of Stacey and Kramers (1975) for initial Pb composition.

Monazite analysis was carried out on SHRIMP II at RSES. Masses analysed are the same as for zircon (Williams 1998), except that mass 202 (Ce phosphate) was used as the reference peak at the beginning of each scan through the masses. Monazite standard USGS44069 was used (424.9 ± 0.4 Ma; Aleinikoff et al. 2006). Energy filtering was used to effectively eliminate isobaric interference on mass 204 (Rubatto et al. 2001), as well as suppressing matrix effects that have been observed in ion probe analysis of monazite (Fletcher et al. 2010). A UO/U versus $^{206}Pb/^{238}U$ calibration, as described by Williams (1998), was used for monazite; the calibration error was between 1–1.5 % (1σ). The fraction of non-radiogenic ^{206}Pb (ƒ206) is typically low for monazite and the main source of non-radiogenic Pb is from surface contamination (CLM-5 and 6 0.4-5 %, Ma9330 1.4-3.2 %, Sp9712 0.5-4.5 %). The ^{207}Pb-correction used a Pb composition of Broken Hill galena, which is the approximate composition of common Pb in the laboratory or the model common Pb composition of Stacey and Kramers (1975) and the choice makes no difference to the age results within uncertainty. U-Pb ages are preferred to Th-Pb age because the high counts on the Th peak can compromise its accurate measurement by electron multiplier during SHRIMP analysis and indeed Th ages are more scattered. Data were reduced using Squid 2 (Ludwig 2009) and Isoplot/Ex (Ludwig 2003). All average and isochron ages are given at 95 % confidence level.

Rutile TIMS U-Pb analysis

Dating rutile by isotope dilution U-Pb method was performed at the SPIDE[2]R lab, RSES, ANU. Hand-picked rutile fractions were cleaned and digested at 190°C in a 4:1 mixture of 25 M HF and concentrated HNO_3 in 3 mL Teflon vials in Parr

Teflon bombs. A mixed U-Pb double spike (^{202}Pb-^{205}Pb-^{233}U-^{236}U) was used as isotopic tracer for rutile unknowns; a ^{205}Pb-^{233}U-^{236}U-^{229}Th spike was used for digestion and column blanks. For the first batch, chemical separation of Pb from rutile followed the established HBr-based protocol (Amelin et al. 2010), where the sample is loaded into the anion exchange column in 0.3M HBr. In the second batch the protocol was modified to use a mixture of 0.3 M HBr + 0.3 M HF was used for the first pass through the columns, in order to assure that Ti stayed in solution and did not precipitate or form colloidal particles. There is no evidence that the different Pb extraction procedures affected the age results. In both batches, samples were loaded into 0.05 ml micro columns packed with Eichrom AG 1x8 200-400 mesh anion exchange resin; Pb was eluted in 0.5 M HNO$_3$. The second pass through the columns followed established HBr-based protocols for Pb separation. Separation of U from the matrix was achieved using UTEVA resin and a mixture of 3 M HNO2 + 1 % H$_2$O$_2$ for bulk washes. U was eluted using 0.02 M HNO$_3$ + 0.2 M HF. Low-U feldspar from the same sample was used for initial Pb correction (composition in Supplementary Table A8b). The established HBr protocol was used for feldspar Pb separation; separation of U was identical to that used for rutile. For the first batch, both U and Pb fractions were loaded onto outgassed Re filaments with Si gel and phosphoric acid for TIMS analysis. The U fractions of the second batch were diluted in 0.2 ml of 0.5 M HNO$_3$ for analysis by MC-ICPMS.

Pb isotopes were analysed using the Triton Plus Thermal Ionisation Mass Spectrometer (TIMS) at the ANU in static multi-collector mode. Analysis of U isotopes was performed on the Triton TIMS in static multicollector mode (1st batch) and Neptune Plus MC-ICPMS (2nd batch). Calculation of Pb and U in rutile

fractions was made using the excel-based AnySpike (Schmitz and Schoene 2007);

data reduction and error propagation of the feldspar fraction was done using

PBDAT (Ludwig 1993). Initial Pb corrections were made using the isotopic

composition of low-U feldspar from the same sample (see below). Synthetic age

standard ET500 (Condon et al. 2008; Huyskens et al. 2016) returned a ^{206}Pb/^{238}U

age of 499.19 ± 0.45 Ma (within 0.2 % of the nominal 500 Ma value).

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Supplementary Tables captions

Table A1: Major and trace element whole rock compositions by XRF (wt %) and LA ICPMS (ppm).

Table A2. Representative garnet major and trace element analyses.

Table A3. Rutile trace element analyses (measured by LA-ICP-MS; internal Standard TiO2=98 wt%).

Table A4. Allanite major and trace element analyses (measured by EDS and WDS - major - and LA-ICP-MS - traces). Concentrations are in oxide wt % for major elements and ppm for trace elements.

Table A5. Monazite major and trace element analyses (measured by LA-ICP-MS). Concentrations are in ppm when not otherwise specified

Table A6. Monazite U-Th-Pb analyses measured by SHRIMP ion microprobe.

Table A7. Allanite U-Th-Pb analyses measured by SHRIMP ion microprobe.

Table A8. Results of TIMS analysis of rutile and description of grain fractions. $^{206}Pb/^{204}Pb$ ratios are corrected for spike and fractionation only. Radiogenic ratios are corrected for spike, fractionation and initial Pb using the K-feldspar composition. Data were reduced and errors propagated using AnySpike (Schmitz and Schoene, 2007).

Table A8b. Measured Pb isotopic composition of low-U-feldspar