1 Revision 1

2	Inherited Eocene magmatic tourmaline captured by the Miocene Himalayan
3	leucogranites
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18	Abstract
19	The Miocene Cuonadong leucogranites, in the easternmost section of the Tethyan
20	Himalaya, Southern Tibet are characterized by two types of tourmaline. Tourmaline
21	occurs as needle-like crystals in the two-mica ± tourmaline granites (Tur G) and large
22	patches in the pegmatites (Tur P). Both the granite and the pegmatites yield Miocene

ages (ca. 20 Ma) based on monazite U(-Th)-Pb dating, whereas ⁴⁰Ar/³⁹Ar 23 geochronology of the coarse-grained tournalines (Tur P) crosscut by pegmatite veins 24 25 yielded an Eocene mini-plateau age of 43 ± 6 Ma. Major element concentrations of 26 tourmaline indicate that both Tur P and Tur G belong to the schorl group with a 27 magmatic origin, but trace elements such as V indicate that they are not cogenetic. 28 Boron isotopes suggest that Tur P (average -9.76‰) was derived from typical crustal sources whereas Tur G (average -7.65‰) contains relatively more mafic input. The 29 30 capture of Eocene tourmaline by the Miocene leucogranites at Cuonadong suggests 31 that the crustally-derived Eocene magmatism may have occurred in the southern Tethyan Himalaya. Identification of the inherited magmatic tourmaline (Tur P), 32 although not common, challenges the current application of tourmaline chemistry to 33 34 the investigation of magmatic-hydrothermal systems.

Keywords: Inherited tourmaline, Himalayan leucogranite, ⁴⁰Ar/³⁹Ar and U(-Th)-Pb
 geochronology, B isotope

37 Introduction

The Himalayan continent-continent collisional belt resulted from the convergence and collision of India and Asia along the Indus-Tsangpo Suture zone that began in the Cenozoic (Yin and Harrison, 2000). Crustal anatexis related to this large-scale continental collision resulted in the formation of a series of leucogranites (Yin et al., 2006), which generally consist of cogenetic two-mica-, tourmaline- and garnet-bearing rocks, with widespread dikes and stocks of pegmatite (Wu et al., 2020). Two sub-parallel E-trending leucogranite belts, the Higher Himalayan and Tethyan

45 Himalayan (Supplementary Figs. 1a, b), have been recognized, with the former exposed along the South Tibetan Detachment System (STDS) in the Higher 46 47 Himalayan Sequence (HHS) and the latter mainly occurring in the core of the North Himalayan Gneiss Domes (NHGDS) (Supplementary Fig. 1b; Wu et al., 2020). The 48 49 majority of the leucogranites have yielded Miocene ages (26-7 Ma), with a small number of samples with Eocene ages (46–30 Ma) being found in the eastern most 50 region of the Tethyan Himalayan (Wu et al., 2020). The Miocene and Eocene 51 leucogranites were proposed to have formed from distinct episodes of crustal anatexis 52 53 with clearly separated distribution in Southern Tibet (Supplementary Fig. 1; Patiño Douce and Harris, 1998; Hou et al., 2012). Tourmaline, which is very common in the 54 55 Himalayan leucogranites and typically the dominant reservoir of B in the rocks, is 56 stable in various P-T-X conditions and could record the physical and chemical conditions under which it formed (Marschall and Jiang, 2011; Slack and Trumbull, 57 2011). Due to its robustness, tourmaline chemistry has recently been used to 58 59 investigate the genesis of Himalayan leucogranites (Yang et al., 2015; Hu et al., 2018). However, these studies relied on the assumption that the tourmalines formed 60 61 cogenetically with their magmatic host rocks as is widely interpreted in most of the global tourmaline occurrences (van Hinsberg et al., 2011). Following the approach 62 illustrated by Thern et al. (2020), we applied the ⁴⁰Ar/³⁹Ar dating method to 63 coarse-grained tourmalines from the Miocene Cuonadong leucogranite, which yielded 64 65 Eocene ages. The identification of inherited tourmalines not only contributes new insights into the Himalayan collisional orogeny, but also provides constraints on 66

67 application of tourmaline chemistry to petrological studies.

68 Cuonadong tourmaline petrography

69 The Cuonadong leucogranite is located in the easternmost section of the Tethyan 70 Himalaya (Supplementary Fig. 1a) and consists mainly of two-mica \pm tourmaline 71 granite and granitic pegmatite. The pegmatites commonly occur as veins or pockets in 72 the leucogranites, without clear boundaries between them (Supplementary Fig. 2). The wall rocks consist mainly of sandstone, mudstone, slate and schist intercalated 73 with carbonates (Li et al., 2017; Zhou et al., 2019). Two types of tourmalines have 74 75 been identified in the Cuonadong leucogranites, large tourmaline crystals in the pegmatites (Tur P; Figs. 1a-d) and needle-like tourmaline crystals in the two-mica \pm 76 77 tourmaline granite (Tur G; Fig. 1e). The Tur P are pervasively distributed in the 78 pegmatites, and are cut by abundant quartz/pegmatite/quartz-muscovite veins along fractures (Figs. 1a-d; Supplementary Fig. 3a). Abundant micro-fractures and some 79 80 zircon inclusions were observed in the Tur P in backscattered electron (BSE) images 81 (Supplementary Fig. 3b). The Tur G occur as disseminated needle-like crystals, 82 coexisting with muscovite, quartz and feldspar in the granites and show clear core-rim 83 zoning in thin section but not in BSE images (Fig. 1e, Supplementary Figs. 2c-d). Both Tur P and Tur G are commonly homogeneous in BSE images. 84

85 Methods

Tourmaline ⁴⁰Ar/³⁹Ar analyses were performed on one pegmatite sample using
an ARGUS VI at the Western Australian Argon Isotope Facility, Curtin University.
Monazite LA ICP-MS U(-Th)-Pb geochronology of both granite and pegmatite were

performed utilizing a system consisting of ASI RESOlution S-155 193nm ArF
Excimer laser coupled to Thermo Scientific iCAP Qc quadrupole ICP-MS at the State
Key Laboratory for Mineral Deposits Research, Nanjing University, China. The
detailed analytical procedures and conditions of all the above and other methods are
listed in Supplementary Appendix A.

94 **Results**

The Tur P in the pegmatites yielded a ${}^{40}\text{Ar}/{}^{39}\text{Ar}$ mini-plateau age of 43 ± 6 Ma 95 (MSWD = 0.8; P = 0.57; Supplementary Table 1; Fig. 2a), which includes 66.3% of 96 the total amount of ³⁹Ar that was released. Commonly for low potassium minerals 97 containing excess argon, a saddle-shaped ³⁹Ar release spectrum during stepped 98 heating will be displayed (Kelley, 2002), which is not shown in our Tur P data 99 indicating the absence of excess ⁴⁰Ar in the lattice during crystallization (Qiu et al., 100 2007), suggesting that the calculated Ar plateau age represents the true age of 101 tourmaline, rather than the result of excess argon. The high apparent step ages (older 102 103 than 70 Ma) displayed within the first few percent of gas released are likely attributed to excess ⁴⁰Ar in fluid inclusions or the margins of the crystals. We calculated a 104 mini-inverse isochron age of 58 ± 15 Ma (MSWD = 0.8; P = 0.63), associated with a 105 trap ${}^{40}\text{Ar}/{}^{36}\text{Ar}$ ratio of 251 ± 52. This ratio, although imprecise, does overlap with 106 atmospheric compositions (~ 298.6; Lee et al., 2006) which would indicate the 107 mini-plateau age (which assumes a ⁴⁰Ar/³⁶Ar ratio of 298.6) is correct. Note that a 108 sub-atmospheric ratio and the absence of saddle-shaped ³⁹Ar release spectrum suggest 109 that no excess ⁴⁰Ar is present in this part of the age spectrum. In addition, some 110

monazite grains in the granite yielded similar ages to the mini-plateau age (Fig. 2b). 111 In general, the mini-plateau and mini-inverse isochron ages of 43 ± 6 Ma and 58 ± 15 112 113 Ma (Fig. 2a), respectively, overlap with each other and are both clearly older than the U-Th-Pb age of ca. 20 Ma for the monazite (Figs. 2b and 2c). 114 Monazite often has excess ²⁰⁶Pb, resulting in high ²⁰⁶Pb/²³⁸U age and reverse 115 discordance on U-Pb diagrams (Schärer, 1984). Thus, the ²⁰⁷Pb/²³⁵U may provide the 116 best estimate of the age. However, the Himalaya leucogranites are too young to yield 117 reliable ²⁰⁷Pb/²³⁵U (Wu et al., 2015). As a result, the Th-Pb ages are often used for 118 119 monazite geochronology of the Himalaya leucogranites (Harrison et al., 1995). Monazite grains from the two-mica \pm tourmaline granite sample yielded a ²⁰⁸Pb/²³²Th 120 age of 20.3 \pm 0.2 Ma, with two grains having older ages of 45.2 \pm 1.6 Ma and 43.9 \pm 121 122 1.4 Ma (Supplementary Table 2; Fig. 2b). The older ages are consistent with the plateau ⁴⁰Ar/³⁹Ar age of the Tur P and represent inherited monazite crystals. The 123 pegmatite sample yielded a monazite 208 Pb/ 232 Th age of 20.5 ± 0.1 Ma 124 (Supplementary Table 2; Fig. 2c), nearly identical to the two-mica granite ages and 125 distinct from the plateau ${}^{40}\text{Ar}/{}^{39}\text{Ar}$ age of Tur P. 126

Major and trace element compositions of the tourmaline are provided in Supplementary Table 3. Both Tur P and Tur G are dominantly schorl end-member tourmalines, with Fe/(Fe+Mg) ratios of 0.67–0.85 for Tur P, 0.75–0.87 for the Tur G cores and 0.74–0.87 the Tur G rims. Tourmaline grains have wide ranges of Ti (0.04–0.15 apfu for the Tur P, 0.01–0.09 apfu for the Tur G core, 0.02–0.11 apfu for the Tur G rim) and Al concentrations (6.23–6.55 apfu for the Tur P, 6.04–6.50 apfu for 133 the Tur G core and 6.00–6.70 apfu for the Tur G rim). On the Al-Fe-Mg diagram, both the Tur P and the Tur G plot in the field of Li-poor granitoids and associated 134 135 pegmatites and aplites, suggesting a magmatic origin (Supplementary Fig. 4). The trace element compositions in tourmaline are commonly low, from tens of ppm for the 136 137 LILE (Large Ion Lithophile Elements) to ppb for the HFSE (High Field Strength Elements). The concentrations of Cr, Co, Ni, Sr, V and Sc in Tur P are commonly 138 higher than those of the Tur G. The Tur P have a wider range and lower average 139 δ^{11} B values than the Tur G, with the former ranging from -14.14‰ to -7.06‰ 140 141 (average -9.76‰) and the latter from -8.82‰ to -6.79‰ (average -7.69‰, Tur G core) and -10.25‰ to -5.83‰ (average -7.61‰, Tur G rim; Fig. 2d), respectively. The 142 δ^{11} B values for Tur P and Tur G are well within the range reported for other granites 143 in the world (Supplementary Table 4; ca. -15% and ca. -5%, Marschall and Jiang, 144 2011). 145

146 Discussion and implications

147 Tournaline is common in highly evolved granites and pegmatites and has been widely studied to investigate their petrogenesis (e.g., Yang et al., 2015) and to 148 149 decipher the process of fluid flow during the magmatic-hydrothermal transition (Launay et al., 2018). All of these studies depend on the assumption that the formation 150 of the tourmaline was coeval with, and has a genetic connection to, the host rocks. 151 Our work at Cuonadong shows that some tourmalines in the granite/pegmatite may be 152 153 inherited and thus unrelated to the host granite. Two lines of evidence support the inherited origin of Tur P: 1) Monazite grains in the pegmatites and two-mica ± 154

tourmaline granites yielded ages of ca. 20 Ma (Fig. 2b), whereas the Tur P 40 Ar/ 39 Ar dating yielded an Eocene age of ca. 40 Ma, coincided with the presence of two Eocene monazite grains in the granite (Figs. 2a, b), and significantly older than the crystallization age of the host pegmatites; 2) The common occurrence of fractures in these tourmalines and the presence of the pegmatite mineral assemblages in the fractures suggests that they predate the pegmatites (Figs. 1a-d).

Several lines of evidence suggest that the two-mica granite and pegmatite 161 evolved in a cogenetic system, with the pegmatite magmas representing the late 162 163 evolved phase: 1) They yield comparable ages; 2) Both the granite and pegmatite have similar mineral assemblages consisting of plagioclase, k-feldspar, muscovite ± 164 165 tourmaline; 3) The occurrences of the pegmatites as veins or pockets in the granites, 166 without clear boundaries between them (Supplementary Fig. 2); 4) Xie et al. (2020) studied zircon grains in the granite and pegmatite at Cuonadong and proposed 167 magmatic evolution from the granites to pegmatites based on the Zr/Hf ratios of 168 169 zircon.

However, Tur P should be xenocrysts in the host granitoids. In a cogenetic system, tourmaline crystallized in early-stage granite should have higher V contents compared to those formed in a late pegmatite because V is preferentially fractionated into early-crystallized ilmenite, biotite and tourmaline during magma crystallization (van Hinsberg, 2011). Therefore, if Tur P is a primary phase in the host pegmatite, it should have lower V contents than Tur G which represents the early crystallization stage (two-mica granite). However, the obviously higher V concentrations in Tur P

177 suggest that it was not a product of the same Miocene magmatism as the host178 pegmatite (Supplementary Table 3).

179 What was the source of the Eocene Tur P and how was it incorporated in the Miocene leucogranite? Based on 40 Ar/ 39 Ar geochronology, the Tur P formed at ca. 40 180 181 Ma. These ages are coeval with the emplacement ages of the Yardoi granitoids (Zeng et al., 2011; Hou et al., 2012), which is ca. 100 km north of Cuonadong and one of the 182 few occurrences of Eocene intrusions in the northern Tethyan Himalaya 183 (Supplementary Fig. 1). The Yardoi granitoids were interpreted to have formed from 184 185 the partial melting of the lower crust of the India plate during Eocene crustal thickening and the subsequent Neo-Tethyan slab break-off in the early stage of the 186 187 India-Asian collision (Fig. 3a; Zeng et al., 2011; Hou et al., 2012). Tourmalines are 188 not common in the Yardoi granitoids but recent boron isotope analyses showed that the δ^{11} B values of the Yardoi granitoids range from -8.9% to -6.6% (Fig. 2c; Hu et al., 189 2018), partially overlapping with those of Tur P having ranges of -14.14‰ to -7.06‰ 190 (average -9.76‰). However, δ^{11} B values for Tur P are relatively lighter, 191 approximating the average boron isotopes of continental crust ($-10 \pm 3\%$, Marschall 192 193 and Jiang, 2011), whereas the Yardoi and the Miocene tourmalines at Cuonadong (Tur G) have higher δ^{11} B values. The differences may suggest more involvement of lower 194 crust (mafic magma) during crustal anatexis for the latter (Fig. 2d), supported by 195 regional tectonic evolution models for the Eocene Yardoi granitoids and Miocene 196 Cuonadong leucogranite (Hou et al., 2012; Wu et al., 2020). We propose that partial 197 melting of upper crustal materials may have occurred in the Cuonadong area during 198

199 Eocene crustal thickening, when shear heating elevated the surface heat flow (Hartz and Podladchikov, 2008), causing the temperature to reach the solidus, generating the 200 201 felsic magmatism and forming Tur P (Figs. 3a). During later geologic events, the Tur P was preserved due to their robustness (Marschall and Jiang, 2011; Slack and 202 203 Trumbull, 2011). The widespread Miocene leucogranite magmatism (including the 204 early-stage two-mica granite and late evolved pegmatite phase), formed either by decompressional melting of the crust (Wu et al., 2020) or extensive long-living shear 205 heating (Whittington et al., 2009; Hou et al., 2012) linked to Indian plate rollback and 206 207 breakoff (DeCelles et al., 2011). The magma ascended to the upper crust (Fig. 3a) where the late pegmatite phase in the upper part of the two-mica granite incorporated 208 209 the early-stage Eocene granitoids and Tur P at shallow levels in the crust (Figs. 3b, c). 210 Although no Eocene tourmaline has been found in the two-mica granite, the presence of Eocene-age monazite xenocrysts (Fig. 2b) are consistent with assimilation of 211 212 Eocene granites by the Miocene granite at Cuonadong.

Our studies document the spatial coexistence of Miocene and Eocene 213 magmatism in the Tethyan Himalaya and suggest petrogenesis through upper crust 214 215 anatexis during shear heating for the Eocene magmas in Southern Tibet. The presence 216 of inherited tourmaline means that the K/Ar clock in the tourmaline was not reset by thermal diffusion during the emplacement of the granitic pegmatite, perhaps due to 217 the high closure temperature and low diffusion rates for major and trace elements 218 within the tourmaline lattice (Dutrow and Henry, 2011). Thus, caution must be used 219 when applying tourmaline geochemistry to the investigation of the petrogenesis of the 220

221 host magmatic rocks.

222 Acknowledgements

- 223 This study was funded by the National Natural Science Foundation of China
- 224 (41725009), and the Fundamental and Applied Fundamental Research Major Program
- of Guangdong Province (2019B030302013). We appreciate comments by Calvin
- 226 Barnes, Nadia Mohammadi and an anonymous reviewer on this paper.

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- **304** Figure captions

Fig. 1 (a) Hand specimen of the Cuonadong pegmatites, showing abundant fractures
in the tourmaline grains. A part of a thin section was scanned as insert, showing the
common occurrences of fractures in Tur P; (b) Another sample of the Cuonadong
pegmatites showing fractured tourmaline; (c) Photomicrograph showing Tur P
crosscut by a quartz vein in pegmatite; (d) Photomicrograph showing Tur P crosscut
by a quartz-muscovite vein in pegmatite; (e) The crystal habit of tourmaline (Tur G)
in the two-mica ± tourmaline granite.

Fig. 2 (a) Plateau ⁴⁰Ar/³⁹Ar dating results, and ³⁶Ar/⁴⁰Ar versus ³⁹Ar/⁴⁰Ar normal and
inverse isochron plots of the Tur P; (b) and (c) Monazite geochronological results of
the Cuonadong leucogranites; (d) Boron isotope compositions of the Tur P and Tur G
and comparison with various boron reservoirs in nature (after Marschall and Jiang,
2011; Hu et al., 2018).

Fig. 3 (a) Schematic regional section showing the Eocene Yardoi granitoids and 317 granitoids at Cuonadong (100 km south to Yardoi) during crustal thickening. The 318 319 large-scale Miocene Himalayan leucogranites due to intense activity of the STD and HHS extrusion emplaced at Cuonadong area. STD: South Tibetan Detachment; MCT 320 = Main Central Thrust; MBT: Main Boundary Thrust; HHS = High Himalayan 321 Sequence; THS = Tethyan Himalayan Sequence; IYS = Indus-Yarlung suture; GTS = 322 Gangdese thrust system; (b) A simplified cartoon for emplacement of the Miocene 323 Cuonadong leucogranites, represented by the two-mica granite and the late cogenetic 324 pegmatite phase, and xenoliths of the Eocene granitic units containing Tur P; (c) 325 Schematic diagram showing the capture of the Eocene Tur P by the Miocene 326

- 327 pegmatites at Cuonadong. Pl = plagioclase; Qz = quartz; Kfs = K-feldspar; Mnz =
- 328 monazite; Ms = muscovite.

Figure 1



Tur P

1 cm

Tur P

Fractúres

Tur P

Qz

300 µm

(b)

-Ms

Qz

Tur P

Fracture

Tur G

(e)

Two mica ± tourmaline granite





Figure 3

(b)



Miocene granitoids





δ¹¹B: average -7.65‰

Tur G

V: average 7.5 ppm

Two-mica ± tourmaline granite