

1 Evolution of the melt source during protracted crustal anatexis;  
2 an example from the Bhutan Himalaya

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20 **ABSTRACT**

21       The chemical compositions of growth zones of magmatic zircon provide  
22 powerful insight into evolving magma compositions due to their ability to record  
23 both time and the local chemical environment. *In situ* U-Pb and Hf isotope  
24 analyses of zircon rims from Tertiary leucogranites of the Bhutan Himalaya  
25 reveal, for the first time, an evolution in melt composition between 32-12 Ma.  
26 The data indicate a broadly stable melt source from 32 Ma to 17 Ma, and the  
27 progressive addition of an older source component to the melt from at least ~17  
28 Ma, and possibly from as early as 21 Ma. Age-corrected  $\epsilon_{\text{Hf}}$  ratios decrease from  
29 between -10 to -15 down to values as low as -23 by 12 Ma. Complementary  
30 whole-rock Nd isotope data corroborate the Hf data, with a progressive decrease  
31 in  $\epsilon_{\text{Nd(T)}}$  from ~18 to 12 Ma. Published zircon and whole-rock Nd data from  
32 different lithotectonic units in the Himalaya suggest a chemical distinction  
33 between the younger Greater Himalayan Series (GHS) and the older Lesser  
34 Himalaya Series (LHS). The time-dependent isotopic evolution shown in the  
35 leucogranites demonstrates a progressive increase in melt contribution from  
36 older lithologies, indicative of increasing LHS involvement in Himalayan melting  
37 over time. The time-resolved data therefore are consistent with a model  
38 wherein LHS material was incorporated into the base of the hanging wall of the  
39 Main Central thrust from ~17 Ma. Exhumation of this hanging wall material  
40 along the thrust triggered decompression melting under fluid-absent conditions  
41 during the later stages of orogenesis.

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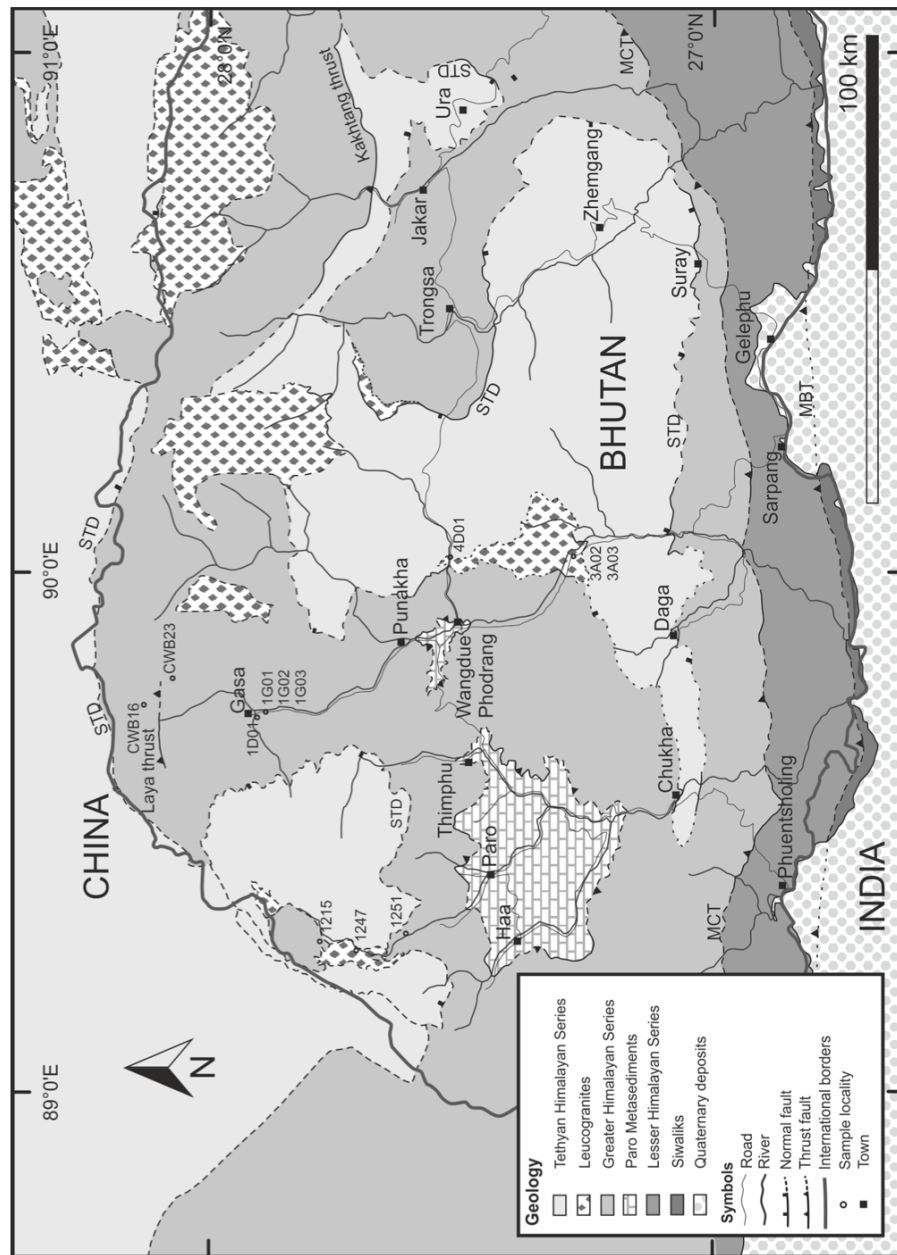
## 44 INTRODUCTION

45 Crustal melting is a fundamental process both for chemical differentiation  
46 and for facilitating ductile deformation of the Earth's continental crust. In the  
47 Himalaya, Late Oligocene-Miocene leucogranites provide a well-documented  
48 example of crustal melting induced by continental collision. Oxygen and hafnium  
49 isotope data from Himalayan leucogranite zircon rims demonstrate that these  
50 granites are examples of pure crustal melting with no detectable mantle input  
51 (Hopkinson et al., 2017). The timeframe across which melting has occurred along  
52 the Himalaya orogen ranges from at least 25 Ma to 9 Ma (Guo and Wilson, 2012),  
53 although granites as old as 39 Ma has been identified in the western Himalaya  
54 (Prince et al., 2001). Despite this extensive period of regional anatexis, no time-  
55 dependant change in the magmatic source has been recognised thus far. The  
56 evolving thermal regime during collisional orogenesis should lead to migration  
57 of melt source regions, and involvement of sources from different crustal levels.  
58 In the case of the Himalayan leucogranites it is possible that the apparent  
59 absence of time-dependent trends results from the lack of chronological  
60 resolution using traditional whole-rock isotopic data. The importance of  
61 recognising the temporal evolution of melt zones lies in better constraining  
62 models of thermal and structural evolution of the continental crust during  
63 orogenesis. Here we evaluate the Hf isotope composition of zircon sampled from  
64 leucogranites in Bhutan (Eastern Himalaya), along with new whole-rock Nd  
65 isotope data. The temporal changes observed in isotopic composition provides,  
66 for the first time, direct evidence for a time-dependant change in the mid-crustal  
67 material undergoing melting during Himalayan crustal thickening.

68

69 **REGIONAL SETTING**

70 We have analysed the isotopic composition of zircon from peraluminous  
71 Oligocene to Miocene leucogranites exposed in the eastern Himalayan orogen in  
72 Bhutan (Fig.1). The granites represent a range of mineralogical types, including  
73 seven two-mica (1G03, 3A03, 4D01, 1247, 1251, CWB16 and CWB23), two  
74 tourmaline-bearing (1G02, 1215), two garnet-bearing (1D01, 3A02) and one  
75 pegmatitic two-mica (1G01) leucogranite. Across the orogen, such granites  
76 intrude upper amphibolite-facies metasediments of primarily Neoproterozoic  
77 source age, the Greater Himalayan Sequence (GHS; Ahmad et al., 2000; Gehrels et  
78 al., 2011). In Bhutan, the leucogranites are emplaced in the uppermost  
79 lithologies of the GHS, close to or crossing the tectonic boundary with the  
80 Tethyan sediments (Greenwood et al., 2016) as is evident from Fig. 1.  
81 Structurally below the GHS, and underthrust along the Main Central thrust  
82 (MCT), is the Lesser Himalayan Sequence (LHS), a primarily Paleoproterozoic-  
83 sourced stack of metasediments (Ahmad et al., 2000; Gehrels et al., 2011). Both  
84 the GHS and LHS comprise a mix of pelitic, orthogneiss, carbonate and quartzite  
85 compositions. The pelitic assemblages are significantly the most melt-fertile, and  
86 therefore provide appropriate source materials for anatectic melts at  
87 temperatures below  $\sim 760^{\circ}\text{C}$  (at pressures equivalent to melting in the mid to  
88 lower crust; Patiño Douce and Johnston, 1991).



89

90 *Figure 1. Geological map of western and central Bhutan, adapted from Greenwood et*

91 *al. (2016), showing sample localities. STD = South Tibetan Detachment, MCT =*

92 *Main Central Thrust, MBT = Main Boundary Thrust, MFT = Main Frontal Thrust.*

93

94 Whole-rock isotope geochemical data from equivalent High Himalayan  
95 leucogranites exposed across the Himalayan orogen suggest that they formed  
96 largely by partial melting of the pelitic lithologies of the GHS into which the  
97 granites intrude (Le Fort et al., 1987; Harris and Massey, 1994; Hopkinson et al.,  
98 2017). However, a whole-rock Sr–Nd isotope study has suggested that a small  
99 contribution from LHS-derived fluids has also contributed to some leucogranite  
100 melts (Guo and Wilson, 2012). Critically, there is no observed temporal control  
101 on the proposed inputs from the LHS. Our new zircon rim U-Pb and age-  
102 corrected  $\epsilon_{\text{Hf}}$  isotope analyses coupled with whole-rock Nd isotope data show  
103 that, at least in Bhutan, there is progressive increase in melt contribution from  
104 the LHS after ~17 Ma.

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## 106 **METHODS**

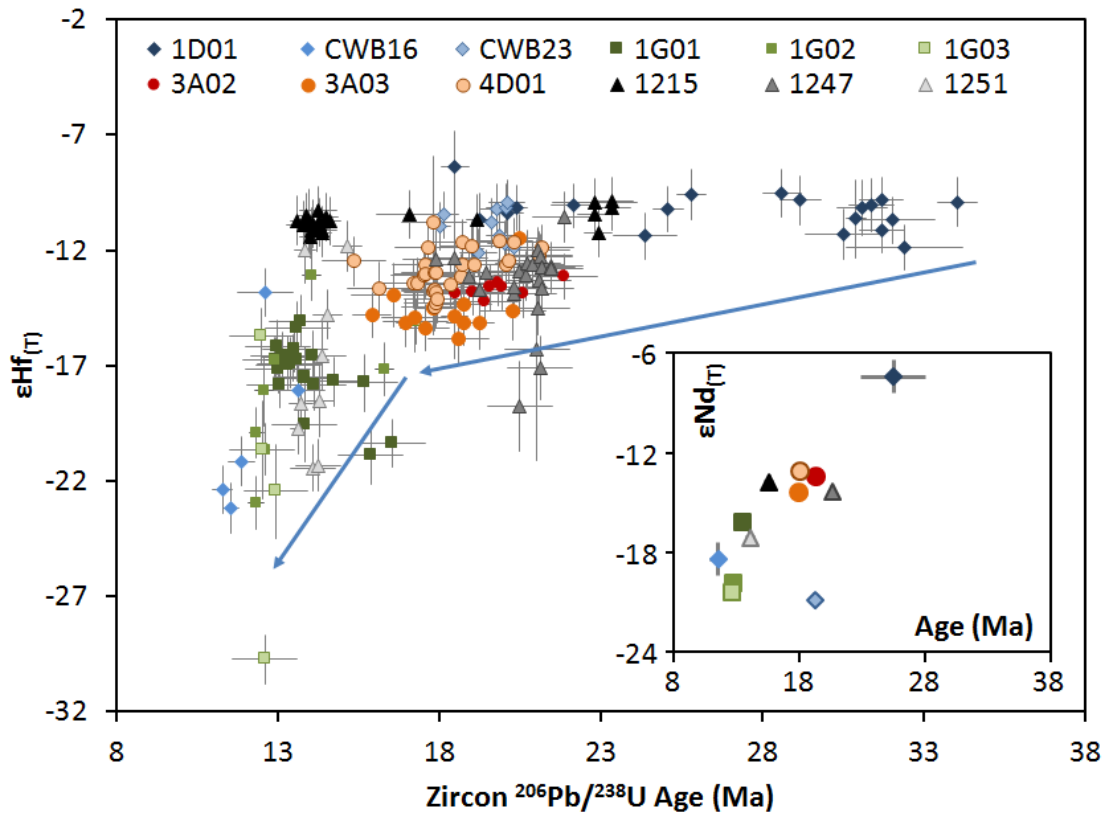
107 Zircon U-Pb and Lu-Hf isotope methods and data were previously reported  
108 in Hopkinson et al. (2017). Whole-rock Sm-Nd isotope analyses were obtained at  
109 the Open University (UK) by thermal ionization mass spectrometry. Further  
110 methodological details and the full dataset is provided in the supplementary  
111 information.

112

## 113 **RESULTS**

114 Zircon from all twelve samples provided rims of Tertiary age ranging from  
115 34 to 11 Ma (Fig. 2, Table DR1), and the majority have inherited cores of Early  
116 Paleozoic or Proterozoic age. The complete dataset is presented in Hopkinson et  
117 al. (2017). Zircon from individual samples record growth over protracted  
118 periods, with spans in individual zircon ages ranging from 0.9 to 15.5 Ma (Fig. 2).

119 This spread of ages from individual hand-samples is typical of Himalayan and  
 120 other S-type granites (Lederer et al., 2013), and the apparent longevity of melt  
 121 formation has been ascribed to protracted timescales of zircon crystallisation  
 122 and long magma residence times (Farina et al., 2018) and more specifically in the  
 123 Himalaya to episodic pulse melting of 1-2 Ma duration (Lederer et al., 2013).



124  
 125 *Figure 2. Zircon rim U–Pb age versus  $\epsilon\text{Hf}_{(T)}$ . Data from Hopkinson et al. (2017) and*  
 126 *Table DR1. Error bars are  $2\sigma$ . Inset shows U–Pb age versus  $\epsilon\text{Nd}_{(T)}$ . Data from Table*  
 127 *DR2.*

128  
 129 Hafnium isotopic compositions are plotted against their corresponding  
 130 zircon age in Fig. 2. Age corrected isotope ratios ( $\epsilon\text{Hf}_{(T)}$ ) range from -8.4 to -29.8.  
 131 Some granite samples preserve large differences in the Hf isotopic composition  
 132 of the zircon rims (e.g. samples 1G03 and 1247). This is likely a result of the lack

133 of homogenisation in the evolving magma, a characteristic that has been  
134 documented in whole-rock studies of Himalayan leucogranites (Deniel et al.,  
135 1987) and may be attributed to high melt viscosities of low-temperature, H<sub>2</sub>O-  
136 undersaturated siliceous magmas (Harris et al., 2000). Variability in Hf isotope  
137 compositions at the hand-sample scale can also be linked to disequilibrium  
138 melting and different mineral phases hosting different reservoirs of radiogenic  
139 Hf (Farina et al., 2014; Tang et al., 2014).

140 Our data show a distinct secular change in  $\epsilon\text{Hf}_{(T)}$  values, with increasingly  
141 lower values in younger samples. Specifically, the zircon compositions show a  
142 change in source, or in the balance between contributing sources, that is initiated  
143 between 21 and 17 Ma. We note that the marked change occurs after 17 Ma, but  
144 outlying data (sample 1247) suggest hints of earlier change at 21 Ma. After 17  
145 Ma, the population of all but one sample (1215) decreases to  $\epsilon\text{Hf}_{(T)} < -16$ . In  
146 terms of two-stage model ages for these zircons, zircon rims older than 17 Ma  
147 have model ages of 1520 to 2000 Ma, whereas those after 17 Ma range from  
148 2000 to 2620 Ma.

149 Whole-rock  $\epsilon\text{Nd}_{(T)}$  ratios range from -7.4 to -20.9 (Fig. 2 inset). There is a  
150 general decreasing trend through time across eleven of the twelve samples. This  
151 trend broadly correlates with the *in-situ* zircon  $\epsilon\text{Hf}$  data, except for sample  
152 CWB23 that has a low  $\epsilon\text{Nd}$  ratio. Assuming that the whole-rock data comprises a  
153 complete dissolution of all minerals, and because the  $\epsilon\text{Hf}$  data fit the broad array  
154 exhibited by all samples, this sample points to a sample-specific disequilibrium  
155 behaviour in the Sm-Nd system.

156

157 **DISCUSSION**

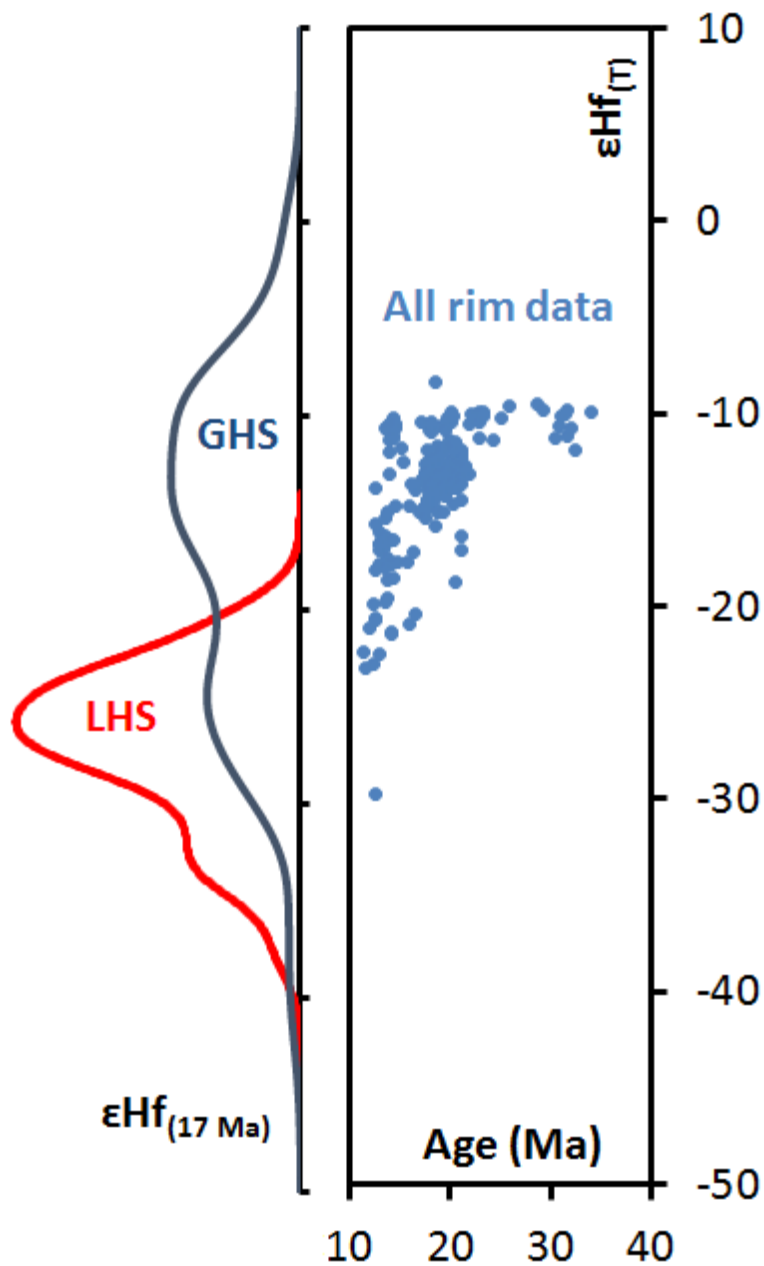


## 158 **An Evolving Magma Source**

159 Previous studies implicate the GHS as the dominant source for Himalayan  
160 leucogranite melts (Harris and Massey, 1994; Guo and Wilson, 2017; Hopkinson  
161 et al., 2017). To address the possible cause of the evolving isotopic signature of  
162 the Bhutanese leucogranites in terms of source rocks, we compare our new  
163 zircon-derived data with existing whole-rock Nd and Hf isotope data from the  
164 Himalayan metasedimentary formations. Previous studies have described a clear  
165 distinction between GHS and LHS units in terms of their isotopic signature, for  
166 example, whole-rock Nd data taken from across the MCT in Sikkim (Mottram et  
167 al., 2014), indicates  $\epsilon\text{Nd}$  values of -12.1 to -18.3 in the GHS (hanging wall), and -  
168 23.4 to -27.7 in the LHS (footwall). These overlap directly with the ranges  
169 obtained from metasedimentary formations in Bhutan of  $\epsilon\text{Nd}$  -12.1 to -17.6 for  
170 the GHS and -25.9 to -32.3 for the LHS (Richards et al., 2006). Whole-rock Nd  
171 isotope data for the samples in this study (Fig. 2, inset) have values of  $\epsilon\text{Nd}_{(T)}$   
172 ranging from -7.4 to -20.9, thus, the entire data array can broadly be generated  
173 from GHS source rocks, but the lower values are compatible with an increasing  
174 LHS component. The recognition of multiple sources for long-duration anatexic  
175 melts is well established in previous zircon studies (Farina et al., 2018).

176 A plot of our *in-situ* zircon Hf isotope data vs. age is shown in Fig. 3 against  
177 data compiled from across the GHS and LHS (Spencer et al., 2018). The compiled  
178 data show that the GHS yields a broad range in  $\epsilon\text{Hf}_{(17)}$  values, from -8 to -30, with  
179 a dominant population between -8 to -18. The LHS mainly falls between  $\epsilon\text{Hf}_{(17)}$  -  
180 21 and -35. A boundary between GHS and LHS could be inferred around  $\epsilon\text{Hf}_{(17)}$  -  
181 22. As with the Nd isotope data, the leucogranite data can be generated by

182 derivation from a GHS source, but also indicate an increasing LHS (or at least  
183 older) source through time.



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185

186 *Figure 3. Zircon rim U-Pb-Hf data from Figure 2 shown with Kernel Density*

187 *Estimates of GHS and LHS detrital zircon data, recalculated at 17 Ma, based on the*

188 *compilation of Spencer et al. (2018).*

189

190 In addition, the dataset of Hopkinson et al. (2017) shows that samples with  
191 1800–1900 Ma zircon cores are only found in samples with younger zircon rims  
192 (Table DR2; Fig. DR2). This Paleoproterozoic orogenic event uniquely affected  
193 the LHS (Ahmad et al., 2000; Kohn et al., 2010), given that the GHS was not  
194 deposited until the Neoproterozoic (Richards et al., 2006; Spencer et al., 2012).  
195 Taken together, the isotopic evidence from zircon rims and whole-rock data, and  
196 the age of zircon cores, strongly suggest that an increasing component of older  
197 and more radiogenic material was being melted during the period of anatexis  
198 from 17 Ma and possibly from as early as 21 Ma.

199 Given that the available melt-fertile (i.e. pelitic) source material is restricted to  
200 two Himalayan formations (LHS and GHS) and that the GHS into which the granites  
201 are emplaced is underthrust by the LHS (Heim and Gansser, 1939), then the results  
202 from this study provide clear evidence for a GHS-sourced melt zone in the Oligocene,  
203 and subsequent tectonic or thermal evolution to allow some melting of the LHS  
204 during the early Miocene (since 17 Ma, and possibly 21 Ma). Direct evidence of  
205 melting in the LHS is yet to be recognised from exposed lithologies in the MCT zone,  
206 but kyanite-grade LHS schists have been identified that have reached temperatures of  
207 ~640°C, sufficient for melting to occur in fertile lithologies (Caddick et al., 2007).

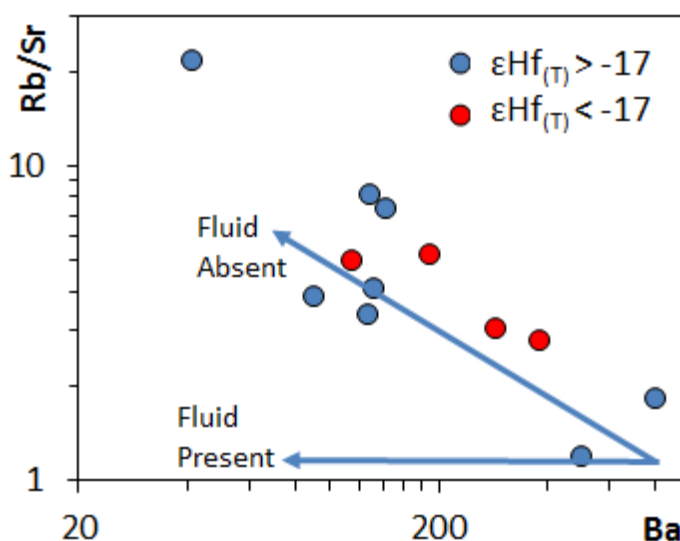
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### 209 **Tectonic Implications**

210 The key finding of this study is that increasing components of older material  
211 are incorporated into the anatectic melt from at least 17 Ma. Whilst the data  
212 presented here do not define a unique tectonic model for the evolution of the  
213 Himalaya, they do require that the pelitic sediments of the LHS are subjected to  
214 melting from the early Miocene during a period of rapid exhumation. For

215 exhumation to cause melting, the melt reaction must be characterised by a  
216 positive gradient of the granite solidus, which in turn requires fluid-absent  
217 melting (Harris and Massey, 1994). The Himalayan leucogranites provide a  
218 classic example of fluid-absent muscovite melting, as indicated by their trace-  
219 element geochemistry (Inger and Harris, 1993); more recently, some examples  
220 of fluid-present melting have also been documented in the Himalaya (Gao et al.,  
221 2017; Huang et al., 2017). The trace-element characteristics of the granites in  
222 this study are indicative of fluid-absent melting (Fig. 4). There is no perceptible  
223 systematic difference in either element chemistry (Table DR3) or mineralogy  
224 between granites with older or younger source regions; each group includes  
225 both peraluminous two-mica and tourmaline-bearing assemblages. These  
226 observations, coupled with empirical thermobarometric evidence for  
227 exhumation of the GHS in the eastern Himalaya at the time of melt formation  
228 ( $\sim 16$  Ma) at a rate of  $2 \pm 1$  mm yr<sup>-1</sup> (Harris et al., 2004), suggest that melting in  
229 Bhutan was induced by decompression during exhumation.

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233

234 *Figure 4. Leucogranite Rb/Sr versus Ba plot. Samples are split by their weighted*  
235 *mean  $\epsilon Hf_{(T)}$  values. Modelled trends are shown for fluid-absent and fluid-present*  
236 *melting (Inger and Harris, 1993).*

237

238 A recent geochemical study of the ~5 km thick MCT zone in Sikkim argued  
239 for progressive tectonic accretion of Lesser Himalayan material from the  
240 footwall to the hanging wall of the MCT as it progressively cut structurally  
241 downwards from ~17 Ma (Mottram et al., 2015; their Fig. 12), a scenario based  
242 on the kinematic model of Bollinger et al. (2006). Metamorphism in the MCT  
243 zone developed as heat advected downward from the overriding GHS  
244 material. Following such accretion, exhumation along the hanging wall of the  
245 thrust could have led to decompression melting of both GHS and accreted LHS  
246 material. The introduction of LHS-sourced material into the melt zone may  
247 therefore mark the initiation of accretion of the LHS into the hanging wall of the  
248 thrust. It is probable that, irrespective of the precise cause of a changing melt  
249 source around the MCT, melting will have enabled continued movement along  
250 this key Himalayan structure through reducing the mechanical strength of  
251 anatexitic rocks around the thrust zone.

252

## 253 **CONCLUSIONS**

254 *In-situ* LA-ICP-MS analyses of zircon rims from Himalayan leucogranites  
255 provide the first evidence for secular compositional change of melts generated  
256 by protracted anatexis during the late Oligocene to mid Miocene. Increasing  
257 components from older source regions from 21-17 Ma to 12 Ma requires

258 increased melting of LHS material during this period. Rapid exhumation of the  
259 hanging wall of the MCT is well documented through the Miocene, and the  
260 granites from the eastern Himalaya provide trace-element characteristics  
261 indicative of fluid-absent melting. A tectonic model that is consistent with all  
262 observations is one in which older LHS material was accreted to the hanging wall  
263 of the MCT, and thus the base of the GHS, during the progressive evolution of the  
264 structure.

265

## 266 **ACKNOWLEDGEMENTS**

267 T.H. acknowledges funding from NERC CASE studentship NE/K501074/1. C.W.  
268 acknowledges funding from NERC Advanced Fellowship NE/HO16279/1. Analytical  
269 work was funded by NERC facility grants IMF478/0513 and IP1403/111. Thanks are  
270 extended to D. Regis and D. Young for field assistance, A. Wood for assistance with  
271 sample preparation at NIGL, and T. Argles, B. Charlier, D. Regis and C. Mottram for  
272 fruitful discussions.

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