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1 Early Earth zircons formed in residual granitic melts produced
2 by tonalite differentiation

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9
10 **ABSTRACT**

11 The oldest geological materials on Earth are Hadean (>4 Ga) detrital zircon grains. Their
12 chemistry and apparently low Ti-in-zircon temperatures ($\leq 700^\circ\text{C}$) are considered to be
13 inconsistent with crystallization in a magma of the tonalite-trondhjemite-granodiorite (TTG)
14 suite, although these are the dominant Archean (4.0–2.5 Ga old) silicic rocks. Using a new
15 dataset of trace element contents in zircons from Paleoproterozoic Barberton TTGs (South Africa)
16 and thermodynamic modelling, we show that these zircons have crystallized at near-solidus
17 conditions from a compositionally uniform granitic melt. This melt is residual from the upper
18 crustal crystallization of a less evolved (tonalitic) parent and thereby shows major and trace
19 element compositions different from bulk TTG rocks. A global compilation reveals that most
20 Hadean detrital and Archean TTG-hosted grains share a peculiar zircon trace element signature
21 that is distinct from the chemical trends defined by Phanerozoic zircons. Our model shows that
22 the low Ti contents of early Earth zircons reflect crystallization at higher temperatures (720–

23 800°C) than initially inferred, due to lower modelled TiO₂ activity in the melt relative to
24 previous estimates. We therefore propose that near-solidus zircon crystallization from a
25 chemically evolved melt in a TTG-like magmatic environment was the dominant zircon-forming
26 process on the early Earth.

27

28 INTRODUCTION

29 The only remnants of the earliest felsic crust are Hadean (4.4–4.0 Ga-old) detrital zircon
30 grains found in younger sedimentary rocks at Jack Hills, Western Australia (e.g. Harrison, 2020).
31 Constraining the nature of the magma in which these zircons formed is key to understanding the
32 geodynamic context of the early Earth. Although rocks of the tonalite-trondhjemite-granodiorite
33 (TTG) suite dominate the earliest preserved Archean (4.0–2.5 Ga old) continental nuclei
34 (Moyen, 2011), they have usually been discounted as potential parents for the Jack Hills zircons
35 (e.g. Harrison, 2020). Specifically, the apparent crystallization temperatures of Jack Hills zircons
36 (≤ 700 °C based on Ti-in-zircon thermometry; Watson and Harrison, 2005) are lower than those
37 of TTG magma formation (> 850 °C), and their trace element compositions do not seem to be in
38 equilibrium with those of TTG bulk rocks (Rollinson, 2008; Reimink et al., 2020; Turner et al.,
39 2020).

40 However, the study of zircon and zircon-hosted inclusions from a 3.81 Ga-old meta-
41 tonalite from the Itsaq Gneiss Complex in Greenland showed that TTG magmas are initially
42 zircon-undersaturated, so zircon should only crystallize at near-solidus conditions in residual
43 granitic melts that formed after extensive cooling and crystallization (Nutman and Hiess, 2009).
44 Moreover, changes in melt chemistry during crystallization may result in zircons forming from a
45 melt showing significantly different trace element compositions than TTG bulk rocks. These

46 possibilities must be systematically addressed to evaluate whether TTG magmas could be
47 plausible parents for Hadean detrital zircons.

48 For this purpose, we investigated the nature and composition of zircon-forming melts in
49 TTG suites, based on new zircon trace element data from the Paleoproterozoic (3.46 Ga-old) TTGs
50 of the Barberton area, Kaapvaal Craton, South Africa (Fig. 1). The compositional evolution of
51 magmas in the course of crystallization, from the tonalitic starting compositions to the near-
52 solidus granitic melts, is well constrained following a recent study (Laurent et al., 2020). The
53 significance of the results is evaluated through a global comparison of Hadean and Archean
54 zircons, based on a compilation of trace element compositions in Jack Hills detrital zircons ($n =$
55 233) and igneous TTG-hosted zircon ($n = 947$) in 44 samples from Archean cratons worldwide
56 (Fig. 1A). Zircon compositions altered by the effects of radiation damage and/or post-magmatic
57 (hydro-)thermal events were filtered out based on chemical and isotopic criteria from both newly
58 acquired and compiled datasets. Details about data acquisition, compilation and filtering are
59 provided in the Supplementary Material.

60

61 **ZIRCON-FORMING MELTS IN ARCHEAN TTG SUITES**

62 The 3.46 Ga Barberton TTGs consist of trondhjemite plutons with minor diorite, tonalite
63 and granite; the plutons are associated with coeval and co-genetic felsic volcanic rocks of the
64 Buck Ridge Volcanic Complex (Laurent et al., 2020) (Fig. 1B). The diverse igneous lithologies
65 have been proposed to result from the differentiation of a parental liquid of tonalitic composition,
66 whereby the compositional range of the plutonic rocks arises from various degrees of crystal
67 accumulation and residual melt loss along the liquid line of descent (Fig. 1C).

68 Zircons from nine samples representative of the Barberton TTG suite show overlapping
69 trace element concentrations (Fig. 2A and Supplementary Material, Fig. S1). In particular, Ti and
70 Hf are powerful monitors of the crystallization environment of zircon, as Ti relates to the
71 temperature and TiO_2 activity of the host melt (Ferry and Watson, 2007) and Hf is a proxy for
72 melt fractionation (Claiborne et al., 2006). Although bulk-rock SiO_2 contents vary from ~60 to
73 76 wt%, zircon Ti and Hf concentrations in the Barberton samples show restricted ranges of 5.1
74 ± 3.2 ppm and 0.95 ± 0.17 wt.% (2 standard deviations [SD], $n = 58$) respectively (Fig. 2A).
75 Similar observations were made from Phanerozoic igneous suites and interpreted as reflecting
76 late crystallization of zircon from a compositionally homogeneous residual melt in all rock types,
77 for instance in the Bogy Plain zoned pluton, southeastern Australia (Ickert et al., 2011). The
78 observed distribution of zircon Ti and Hf concentrations in the Paleoproterozoic Barberton TTG
79 plutons suggests a comparable situation.

80 The inversion of zircon compositions in the Barberton samples, using the
81 parameterization of zircon/melt partition coefficients as a function of Ti concentration of
82 Claiborne et al. (2018), further supports this interpretation. The composition of the calculated
83 melts indeed overlaps with that of the high-silica granitic melt inclusions from the BRVC (Fig.
84 2B), which represent the most differentiated, residual melts from crystallization of the parent
85 tonalitic magma (Laurent et al., 2020). Importantly, both the calculated melts and BRVC melt
86 inclusions share the same distinctive differences with average TTGs, namely strikingly higher Th
87 and U contents and a lower middle- to heavy rare earth element ratio (Fig. 2B). These features,
88 which likely reflect the highly evolved nature of the melts and significant amphibole
89 fractionation, explain the apparent disequilibrium between trace element concentrations of zircon
90 and the composition of their host TTG rocks (Reimink et al., 2020).

91 We simulated the crystallization of the parental liquid of the Barberton TTG suite (as
92 determined by Laurent et al., 2020) at 3 kbar, using the mineral and melt models of Holland et al.
93 (2018) and the PERPLE_X software program (<https://www.perplex.ethz.ch/>; Connolly, 2009).
94 The phase equilibria and thermodynamic data obtained served to model the evolution of melt Zr
95 concentration as well as zircon saturation temperatures and Ti contents (see Supplementary
96 Material for details). The results show a first step (from the liquidus at 1050 °C to ~40 wt.%
97 liquid at 725 °C) of dominant plagioclase (~40 wt.%), subordinate amphibole (~15 wt.%), and
98 minor pyroxene and Fe-Ti oxide crystallization (Fig. 3A). The melt fraction then decreases from
99 725 °C to the solidus (~700 °C), corresponding to the near-eutectic crystallization of quartz,
100 plagioclase, K-feldspar and biotite. These results match the modal compositions recorded by
101 natural samples (Laurent et al., 2020) and the experimental results of upper crustal tonalite
102 crystallization (Marxer and Ulmer, 2019).

103 The low initial Zr content of the system (50–200 ppm), combined with buffering of the
104 melt Zr concentration by amphibole crystallization, result in a range of zircon saturation
105 temperatures (considering model uncertainties) between 800 °C and 720 °C (Fig. 3A). Zircon
106 crystallizes in a high-silica residual melt similar to the BRVC melt inclusions in both major (Fig.
107 1C) and trace elements (Fig. 2B), which agrees with the observation of zircon in the interstitial
108 domains between plagioclase and hornblende phenocrysts in natural samples (Laurent et al.,
109 2020). These results are consistent with the inferred zircon crystallization temperatures (700–
110 800°C) and granitic mineralogy of zircon-hosted inclusions in a 3.81 Ga-old tonalite from
111 Greenland (Nutman and Hiess, 2009).

112

113 **ZIRCON CRYSTALLIZATION CONDITIONS ON THE EARLY EARTH**

114 Most TTG-hosted igneous zircons and many Jack Hills detrital zircons from our
115 compilation compositionally overlap with the Paleoproterozoic Barberton zircons investigated here
116 (Fig. 2A, 4 and Supplementary Material, Fig. S1). In Figure 4, we compared their Ti and Hf
117 contents to those of zircon from various Phanerozoic igneous systems. Phanerozoic zircons show
118 a negative correlation between Ti and Hf contents (Fig. 4) that reflects crystallization during
119 magma cooling and differentiation (Claiborne et al., 2006). Such correlation may be recorded by
120 zircons from single hand samples, as illustrated by one monzonite and one granite from the
121 Miocene Spirit Mountain Batholith, southern Nevada, USA (Claiborne et al., 2006) (Fig. 4). In
122 turn, distinct parental melt compositions and magma evolution pathways result in distinct zircon
123 Ti-Hf co-variations for different geodynamic settings (Fig. 4).

124 In contrast, most early Earth zircons define a tight compositional cluster in the Ti-Hf
125 space (Fig. 4) with very low Ti contents (2–9 ppm) (Carley et al., 2014) at moderate Hf
126 concentrations (0.8–1.2 wt%). Strikingly, the variations in zircon Ti and Hf contents in the >40
127 early Earth samples taken together are more restricted than in single samples of the Spirit
128 Mountain Batholith (Fig. 4). Although the early Earth zircons partly overlap with the
129 compositionally most evolved, modern convergent margin and post-orogenic zircons, they
130 clearly lack the high Ti (>15 ppm) and/or low Hf (<0.7 wt%) parts of the Phanerozoic trends
131 (Fig. 4). Altogether, this supports the existence of specific conditions of zircon growth for
132 Hadean and Archean zircons. As defined based on the Barberton case, these conditions are the
133 crystallization from a near-solidus, residual granitic melt resulting from differentiation of a
134 tonalitic parent magma, with little (if any) zircon fractionation having occurred beforehand. This
135 condition may reflect the generally lower Zr concentrations of early Earth magmas (<150 ppm)

136 relative to Phanerozoic magmas (Keller et al., 2017), which would delay zircon saturation to
137 near-solidus temperatures.

138 This is confirmed by our crystallization model, in which zircon saturates only at ≤ 800 °C
139 and accordingly shows the low Ti concentrations (<10 ppm) (Fig. 3A-B) typical of early Earth
140 grains (Fig. 2A, Fig. 4). The modelled cumulative curve of Ti contents in zircon predicts a
141 greater proportion of low-Ti zircons than observed in nature (Fig. 3B). This may reflect that
142 during analysis of natural grains, thin zircon rims crystallized at low temperatures were
143 undersampled relative to large, higher-temperature cores (Ickert et al., 2011). However, the
144 observed concentration range is exactly the same (2–9 ppm Ti; Fig. 3B). This is a robust
145 indicator of the model's validity, as this range is controlled by zircon saturation temperature and
146 TiO_2 activity, which depend only on the initial Zr content of the liquid and phase relations during
147 crystallization. Importantly, the model indicates that the low Ti concentrations of the Jack Hills
148 zircons can be explained by crystallization at significantly higher temperatures (720–800 °C; Fig.
149 3B) than have been estimated so far (650–720 °C), because the TiO_2 activity of the system was
150 much lower (ca. 0.4–0.5 as modelled here) than was previously assumed for Ti-in-zircon
151 thermometry (up to 1; Watson and Harrison, 2005).

152

153 **CONCLUSIONS AND IMPLICATIONS**

154 We propose that (1) zircon in TTG suites forms at near-solidus temperatures (720–800
155 °C) from a residual granitic melt resulting from extensive crystallization of a tonalitic parent and,
156 hence, compositionally different from the bulk rocks; and (2) this zircon-forming mechanism is
157 common to all early Earth zircons, including the Jack Hills detrital grains, as shown by their
158 shared peculiar trace element signature that is distinct from that of Phanerozoic zircon. This

159 model reconciles the paradoxical observations that the low-Ti nature of the Jack Hills zircons
160 (Watson and Harrison, 2005) reflects crystallization in a melt close to the granite minimum,
161 although such granites are scarce in the early Earth (Moyen, 2011). This liquid may in fact be
162 similar to the most differentiated melt of the TTG suites, which remained trapped in the
163 crystalline framework and/or is not preserved as co-magmatic silicic volcanic rocks (Laurent et
164 al., 2020).

165 Whether the Jack Hills zircons formed in voluminous TTG plutons, as observed in
166 Archean cratons, or in lower-volume felsic veins and pods in a dominantly mafic crust (e.g.
167 Rollinson, 2008) is still uncertain. In fact, the parameters controlling zircon chemistry, namely
168 the petrogenesis of felsic magmas (intracrustal differentiation of hydrous mafic material) and
169 hence their phase relationships upon crystallization, would be comparable in both cases.
170 Nevertheless, closed-system crystallization as modelled here, which is more likely to occur in
171 small, isolated melt pockets, may also explain the distributions of zircon Ti contents skewed to
172 lower concentrations as compared to crystallization in large, open-system plutons (Fig. 3C).
173 Interestingly, the compositional range of the Jack Hills zircons extends to lower Ti (1–3 ppm)
174 and/or higher Hf (1.2–1.4 wt%) than that of the TTG-hosted zircons (Fig. 4), which may be
175 explained by the smaller amounts of silicic melt and closed-system conditions, and/or even lower
176 initial Zr contents, in the Hadean. Either way, our results indicate that the trace element
177 composition of Hadean detrital zircons is consistent with a TTG-like magmatic environment.

178

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184

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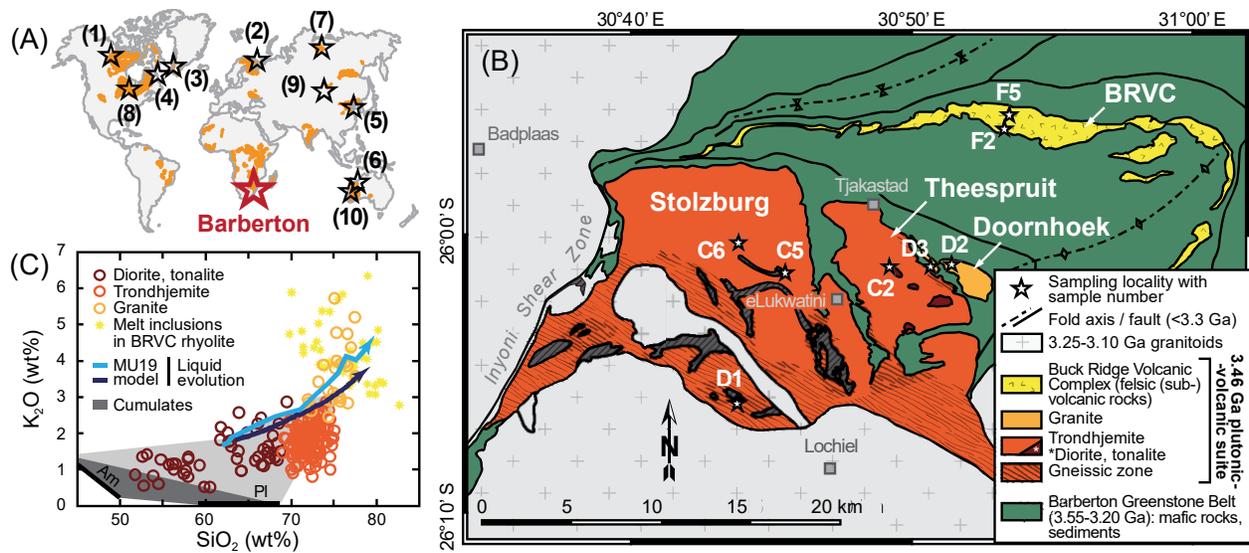
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295

296 **FIGURE CAPTIONS**



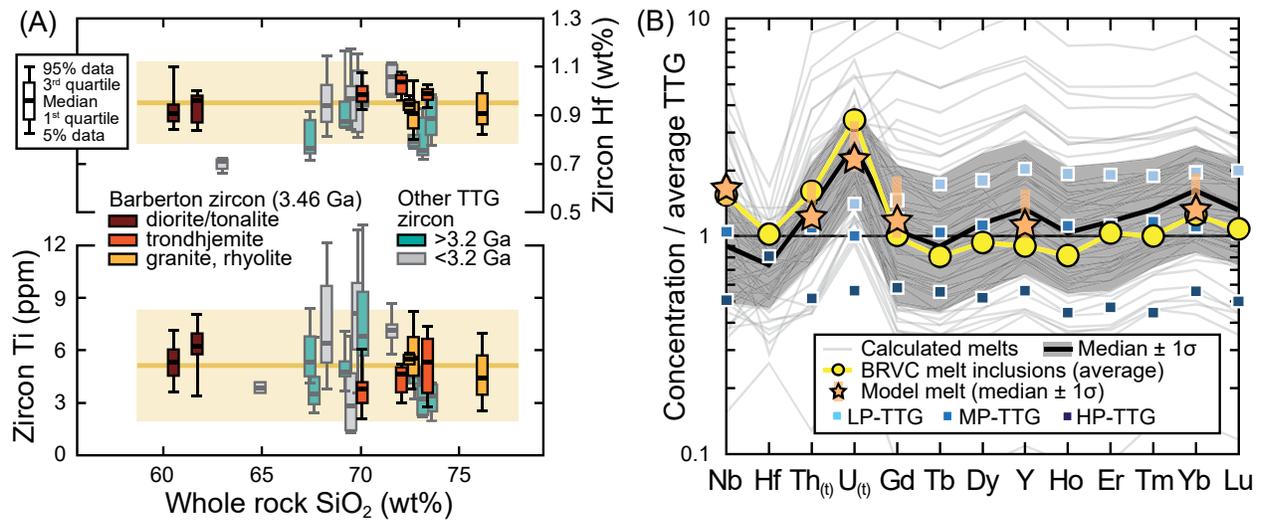
297

298 **Figure 1.** A: Location of zircon trace element data examined here: Barberton (this study); (1)
 299 Acasta Gneiss Complex (Reimink et al., 2020); (2) Kola (Vetrin et al., 2016); (3) Itsaq Gneiss
 300 Complex (Whitehouse and Kamber, 2002; Nutman et al., 2007; Kielman et al., 2018); (4) Uivak
 301 Gneiss (Vezinet et al., 2018); (5) Jiaobei Province (Liu et al., 2013; Shan et al., 2015); (6) North
 302 Pole Dome (Kitajima et al., 2012); (7) Irkut (Turkina et al., 2012); (8) Superior Province
 303 (Bouvier et al., 2012); (9) Tarim (Zong et al., 2013); (10) Jack Hills (Cavosie et al., 2006; Bell et
 304 al., 2014; Turner et al., 2020). B: Geological map of the Barberton Paleoproterozoic plutonic-
 305 volcanic suite, South Africa (Laurent et al., 2020); BRVC = Buck Ridge Volcanic Complex. C:
 306 K₂O vs. SiO₂ composition of bulk rock samples from the Barberton suite, BRVC melt inclusions
 307 and tonalite liquid lines of descent (experimental from Marxer and Ulmer, 2019 and modelled
 308 here – see text); the gray field represents mixed compositions between melts and plagioclase (Pl)
 309 + amphibole (Am) cumulates (adapted from Laurent et al., 2020).

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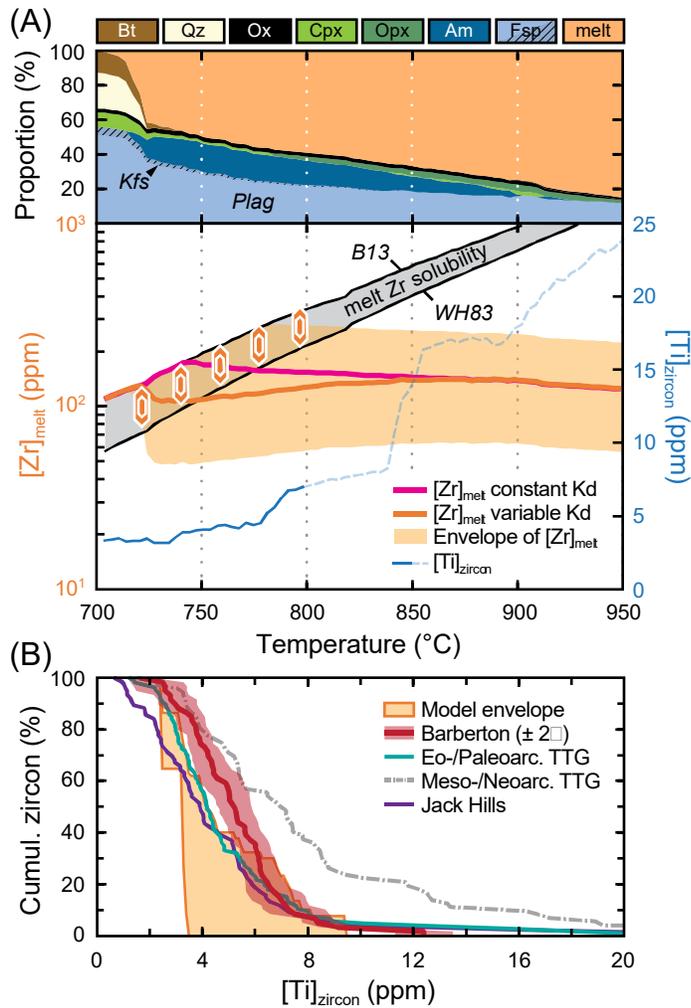
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 314 **Figure 2.** A. Plot of the distribution of zircon Ti and Hf contents in the Paleoproterozoic Barberton
 315 and other Archean TTGs, as a function of the host rock SiO₂ content (one box-and-whisker per
 316 sample). The horizontal lines and bands represent averages ± 2 SD of all Barberton zircons. B.
 317 Trace element concentrations of zircon-forming melts (normalized to average TTG from Moyen,
 318 2011) in the Barberton samples obtained from inversion of zircon compositions, compared to
 319 those of the BRVC melt inclusions; the low-, medium- and high-pressure (LP-, MP-, HP-)TTG
 320 groups (Moyen, 2011); and the melts saturating zircon in the crystallization model (see text). All
 321 Th_(t) and U_(t) concentrations (including normalization values) are corrected for radioactive decay.

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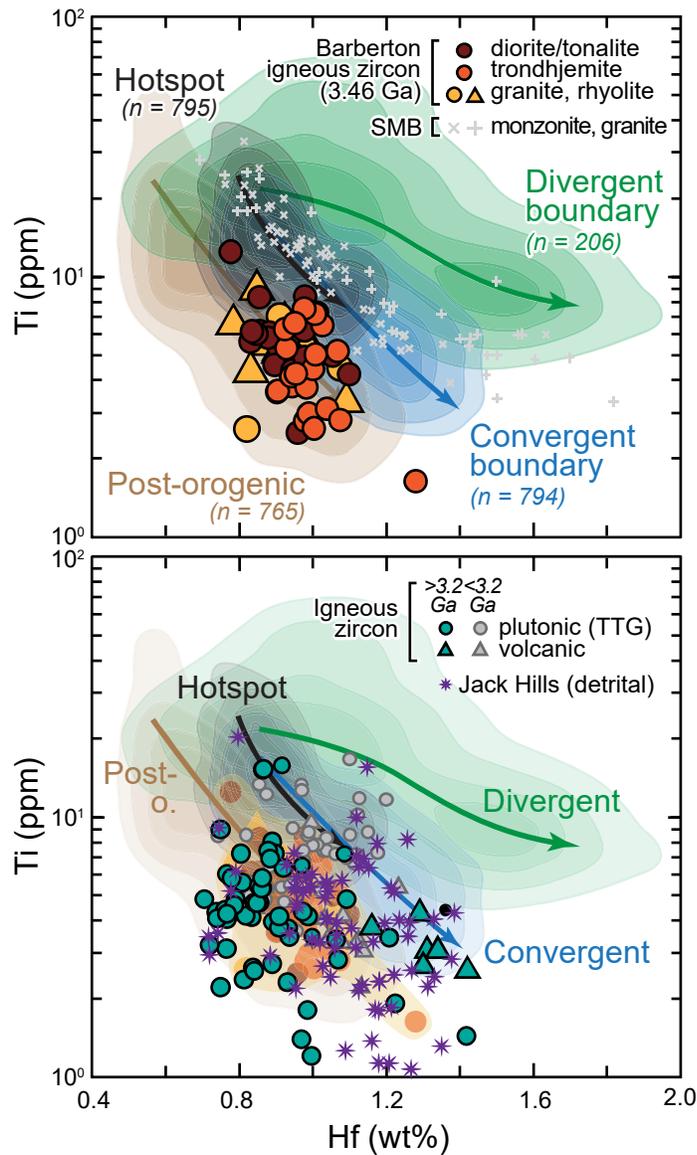
325 **Figure 3.** Results of the tonalite crystallization and zircon saturation model. A. Temperature
 326 evolution of phase proportions (top plot; Bt = biotite; Qz = quartz; Ox = Fe-Ti oxides; Cpx =
 327 clinopyroxene; Opx = orthopyroxene; Am = amphibole; Fsp = feldspar, including plagioclase
 328 [Plag] and K-feldspar [Kfs]); and modelled melt Zr content ($[Zr]_{melt}$, left axis) and zircon Ti
 329 concentrations ($[Ti]_{zircon}$, right axis) (bottom plot). Initial $[Zr]_{melt}$ ranges from 50 to 199 ppm
 330 (bounding the model envelope) with an average of 111 ppm (bold curves, obtained using either
 331 constant or variable Kd for Zr) (see details in Supplementary Material). The dashed curve shows
 332 theoretical $[Ti]_{zircon}$ at temperatures above zircon saturation. The melt Zr solubility range is
 333 bound by the models of Watson and Harrison (1983) (WH83) and Boehnke et al. (2013) (B13).

334 Zircon symbols represent the possible range of zircon saturation conditions within uncertainties.

335 B. Cumulative proportions of $[\text{Ti}]_{\text{zircon}}$ in the model compared to early Earth TTG and Jack Hills

336 zircons.

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340 **Figure 4.** Titanium vs. Hf plots showing the composition of early Earth zircons and Phanerozoic
 341 zircons from different geodynamic settings (data from Burnham and Berry, 2017; Carley et al.,
 342 2014; Grimes et al., 2009; Karakas et al., 2019) as kernel density estimates contoured for 25, 50,
 343 75 and 90% data; the arrows approximate the highest point density trends and correspond to the
 344 zircon compositional evolution throughout magma differentiation from mafic-intermediate to

345 felsic. The crosses depict zircon data from two samples (one monzonite, one granite) of the Spirit
346 Mountain Batholith (SMB) (Claiborne et al., 2006).

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349 ¹GSA Data Repository item 202Xxxx, containing details about analytical and modelling

350 methods, data compilation, filtering, and zircon/melt partitioning, is available online at

351 www.geosociety.org/pubs/ft20XX.htm.