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1 Zircon U-Pb geochronology and Hf isotopes of the Luís Alves Terrane: Archean to Paleoproterozoic 2 evolution and Neoproterozoic overprint

3 Beatrix M. Heller^{1,2}, Mathias Hueck¹, Claudia R. Passarelli¹, Miguel A. S. Basei¹

4 ¹Instituto de Geociências, Universidade de São Paulo, São Paulo, Brazil

5 ²Université Paris-Saclay, CNRS, GEOPS, 91405, Orsay, France

6

7 Abstract

8 The Luís Alves Terrane in southern Brazil is one of the largest expositions of the Archean to
9 Paleoproterozoic units that acted as basement for the development of the Neoproterozoic Pan-
10 African/Brasiliano orogenic belts in the Mantiqueira Province, in South America. Combined field
11 observations, petrography, Hf and U-Pb LA-ICP-MS zircon and titanite isotopic data are presented for
12 the basement of this terrane. The zircon U-Pb dataset spans a large time range with concordant ages
13 from 3.2-1.8 Ga. Several magmatic and metamorphic events are recorded by the U-Pb concordant
14 ages reflecting the complex evolution of this crustal block. The oldest inherited zircon grains have
15 Mesoarchean ages (3.2-3.1 Ga), while two other magmatic populations yield ages of $2,683\pm 70$ Ma
16 and $2,498\pm 11$ Ma. Most zircon grains crystallized during two high-grade metamorphic events at
17 $2,352\pm 23$ Ma and $2,183\pm 17$ Ma. Titanite from two samples crystallized at 2.02-1.99 Ga, probably
18 during a retrograde metamorphic trajectory that re-equilibrated the studied samples in amphibolite-
19 facies conditions. A late pegmatite dyke was emplaced between ca. 1.96 and 1.79 Ga.
20 Cathodoluminescence images show that the analyzed zircon samples, including those in the
21 pegmatite, had their internal structure modified by hydrothermal processes. The localization of the
22 outcrop close to the rim of the Campo Alegre Basin affected by an important Neoproterozoic
23 hydrothermal event as well as normal and reverse discordant U-Pb ages in our dataset indicate that
24 this event took place during the Brasiliano orogenic cycle at the end of the Neoproterozoic. The Hf
25 isotope data show predominantly crustal signatures with $\epsilon\text{Hf}(t)$ values ranging from 1.3 to -16.8. Hf
26 T_{DM} model ages cluster in two groups (4.32-3.48 and 3.38-2.27 Ga) indicating two pulses of magma
27 differentiation from the mantle with subsequent mixing of the material during the Paleoproterozoic
28 high grade metamorphic events.

29 Keywords: Luís Alves Terrane, U-Pb geochronology, Zircon Lu-Hf isotopy, hydrothermal zircon.

30 1. Introduction

31 Zircon crystals from granulitic terranes commonly have complex textures recording multiple
32 crystallization events that can be associated to both magmatism and high-grade metamorphism
33 (Corfu 2003; Hoskin and Black 2000; Kröner et al. 2014; Zhao et al. 2015; Oriolo et al. 2016). A
34 common consequence in HT to UHT terranes is the expanded spreading of U-Pb zircon data along the
35 Concordia or bordering a sub-parallel Discordia line whose meaning requires careful interpretation.
36 The spread of zircon ages may be attributed to a sequence of metamorphic events with episodic
37 zircon growth or partial resetting (Whitehouse and Kemp 2010; Taylor et al. 2016; Rubatto 2017;
38 Laurent et al. 2018; Tedeschi et al. 2018). Additionally, the implications for U-Pb geochronology are
39 amplified in ancient geological terranes, in which the metamictization of zircon crystals results in a
40 higher susceptibility to hydrothermal processes (Geisler et al. 2001). In this context, late opening of
41 the isotopic system may result in complex datasets involving both normal and reverse discordance
42 (Williams et al. 1984; Corfu 2013; Kusiak et al. 2013; Wiemer et al. 2017).

43 The Luis Alves Terrane (LAT), which is ca. 230km long and 50km wide is located in southern Brazil and
44 is surrounded by orogenic belts formed during the Brasiliano / Panafrican orogenic cycle at the end
45 of the Neoproterozoic and Cambrian (Basei et al. 1998a, 2009a, b; Passarelli et al. 2018; Siegesmund
46 et al. 2018). Nonetheless, Neoproterozoic-Cambrian activity in the LAT was restricted to the
47 emplacement of intrusive rocks (Kaul 1984; Gualda and Vlach 2007 a,b) and the development of
48 (volcano-) sedimentary basins (Basei et al 1998b; Rostirolla et al. 1999; Citroni et al. 2001; Guadagnin
49 et al. 2010; Basei et al. 2011b; Quiroz-Valle et al. 2019) while the crystalline basement, the Santa
50 Catarina Granulitic Complex (SCGC), remained largely unaffected (Hartmann et al. 1979, 2000; Basei
51 et al. 1992, 1998a; 2009b, Siga Jr. 1995; Harara et al., 2003; Passarelli et al 2018). All high-
52 temperature geochronological data of the SCGC yield Paleoproterozoic and Archean ages (Basei
53 1985; Siga Jr 1995; Hartmann et al. 2000; Harara 2001; Basei et al 2009 a,b) and the only signs of
54 reactivation of the basement during the Brasiliano orogenic cycle are ~ 600 Ma old K-Ar ages in shear
55 zones (Siga Jr 1995).

56 In this study we present U-Pb LA-ICP-MS ages from zircon and titanite from one basement outcrop in
57 the central-western portion of the LAT. Our data provide the required complement to the available
58 geochronological data set which was mainly obtained from multi-crystal ID-TIMS zircon U-Pb ages
59 and imprecise Rb-Sr whole rock ages .The new data confirm the suggested geochronological
60 evolution and provide more precise age constraints for the main magmatic and metamorphic events
61 that formed the LAT. Detailed evaluation of this complex dataset allows the recognition of a partial
62 opening of the U-Pb system, probably during an hydrothermal event in the late Neoproterozoic
63 Brasiliano orogenic cycle. This event led to redistribution of radiogenic Pb within and out of zircon
64 crystals, resulting in a high number of normal and reverse discordant U-Pb ages. Furthermore, we
65 present Hf isotope data for the LAT, which indicate that the LAT is composed of old crustal material
66 extracted from the mantle in two main pulses which became progressively mixed during tectonic
67 reworking in the Paleoproterozoic.

68

69 **2. Geological setting**

70 Southern Brazil is structurally marked by the juxtaposition of several geotectonic domains with
71 different evolutions (Fig. 1). Neoproterozoic subduction and collision events led to the formation of
72 Western Gondwana during the Brasiliano/Pan-African orogenic cycle affecting the crustal blocks to
73 differing degrees (Heilbron et al. 2004; Basei et al. 2010; Passarelli et al. 2011; Oriolo et al. 2017;
74 Siegesmund et al. 2018). The oldest domains are the Rio de la Plata and Paranapanema cratons,
75 located to the west and presently mostly covered by the sediments of the Paraná Basin. The eastern
76 border of these cratons are marked by two low- to medium-grade metamorphic metavolcano-
77 sedimentary complexes and intrusive granitoids: the Ribeira Belt in the north and the Dom Feliciano
78 Belt in the south. These tectonic domains are separated by two granite-gneiss terranes: the Curitiba
79 Terrane, strongly migmatized in the Brasiliano orogenic cycle and the Luis Alves Terrane (LAT).

80 The Curitiba Terrane is located immediately to the South of the Ribeira Belt, separated by the
81 Lancinha-Cubatão-Itariri Shear Zone (Passarelli et al. 2011). The crystalline basement is 60 km wide
82 and 200 km long and contains the Atuba-Registro-Itatins Complex (Fig. 1). The basement is composed
83 of banded migmatitic gneisses of Paleoproterozoic age (2.1–2.0 Ga) (Siga Jr 1995; Basei et al 1998a)
84 that underwent intense deformation and migmatization during the Neoproterozoic. Features from a
85 second Neoproterozoic migmatization phase occur commonly, associated with the Brasiliano/Pan-
86 African orogenic cycle. The complex is partly covered by low-grade Neoproterozoic metavolcano-
87 sedimentary sequences and was intruded by anorogenic alkaline–peralkaline granitoids of the late
88 Neoproterozoic Serra do Mar Suite, commonly included in the alkaline Graciosa Province (Gualda and

89 Vlach 2007a,b; Vlach et al. 2011). The contact between the Curitiba Terrane and the LAT is made up
90 of calc-alkaline granitoids of the Rio Piên Batholith as well as rocks of the Piên mafic ultramafic suite,
91 both of which were heterogeneously deformed along the Piên and Mandirituba-Piraquara Shear
92 Zone (Machiavelli et al. 1993; Harara 1996). The association is interpreted as remains of a
93 dismembered ophiolitic complex (Basei et al. 1992; Machiavelli et al. 1993; Harara 2001).

94 The LAT is limited to the south by the Itajaí-Perimbó Shear Zone, which separates it from the Brusque
95 Group and the Itajaí Basin, domains of the Dom Feliciano Belt (Fig. 1), which extends for over 1,400
96 km until Uruguay and was formed in the Neoproterozoic by the tectonic interaction between the Río
97 de la Plata, Congo and Kalahari Cratons (Basei et al. 2000; Oyhantçabal et al. 2009; Philipp et al.
98 2016; Oriolo et al. 2016; Hueck et al. 2018). The Brusque Group is a poly-deformed metavolcano-
99 sedimentary sequence that reached upper greenschist to lower amphibolite metamorphic facies
100 (Philipp et al. 2004; Basei et al. 2011a, 2020) and was intruded at ca. 600 ± 15 Ma by voluminous
101 granitic magmatism (Florisbal et al. 2012a, b; Hueck et al. 2016, 2020). Its crystalline basement is
102 exposed in the Camboriú Complex, an association of gneiss-migmatitic rocks and granitic intrusions
103 strongly reworked in the Brasiliano/Pan-African orogenic cycle (Bitencourt and Nardi 2004; Basei et
104 al. 2013; Martini et al. 2015, 2019).

105 Lastly, the eastern termination of the Luís Alves Craton is the Costal Terrane, represented by the
106 Paranaguá Domain. This igneous domain of Neoproterozoic origin (616-590 Ma) comprises a great
107 variety of granitic rocks, most of them with calc-alkaline, magmatic arc signature. Metamorphic rocks
108 such as gneisses, mica-schists quartzites and amphibolites occur as roof pendants within these
109 granitoids (Basei et al. 1990; Siga Jr. 1995; Passarelli et al. 2004; Cury et al. 2008).

110 2.1. The Luis Alves Terrane

111 The LAT (Hartmann et al. 1979, 1998, 2000; Basei et al. 1992, 1998a, 2009b; Siga Jr. 1995; Harara et
112 al. 2002, 2003; Passarelli et al. 2018) consists of an Paleoproterozoic association with Archean
113 protholiths, which in the Neoproterozoic was intruded by granites and locally covered by
114 metavolcano-sedimentary basins. The basement units are grouped into the Santa Catarina Granulitic
115 Complex (SCGC), predominantly a migmatitic–granitic–gneissic association with TTG signatures, while
116 subordinately mafic and metasedimentary units also occur.

117 The southern portion of the LAT is dominated by biotite-amphibole gneiss with biotite-bearing
118 mesosome and pink leucosome. Porphyritic biotite granites and pink mylonitic leucogranites are also
119 common. More seldom are charnockitic–enderbitic rocks and lenticular meta–gabbro bodies in the
120 gneisses forming strongly foliated amphibolites. The main structure is the gneissic foliation with an
121 average orientation is of N30°E/51°NW. The central part of the terrane is dominated by grey
122 charnockitic–enderbitic coarse–grained gneisses with a marked gneissic foliation and numerous
123 enclaves and boudins of amphibolitic mafic rocks. Opx–bearing pegmatite veins cross-cut the gneissic
124 foliation, which may indicate that the thermal apex postdated the main deformational event. Along
125 the South Atlantic coast in the eastern portion of the LAT, the Barra Velha mafic–ultramafic Complex
126 is boudinaged in the felsic granulitic gneisses and contains gabbro, gabbro–norite, amphibolite and
127 websterite, thoroughly recrystallized by high–grade metamorphism (Minioli 1972; Fornari 1998). The
128 northwestern region is dominated by greenish–grey amphibole–biotite–rich orthogneisses
129 containing relicts of orthopyroxene, as well as mafic granulites with associated charnockitic-
130 enderbitic portions. Lenses of amphibolitic schists, serpentinites, garnet–rich amphibolites,
131 amphibolitic gneisses and felsic granulites occur locally. The predominant trend of the structures is
132 NW–SE. In the northernmost portion of the LAT, leucocratic felsic tonalitic to granodioritic gneisses
133 with many intercalations of greenish charnockite layers prevail, commonly with mylonitic portions.

134 Hyperstene in the orthogneisses indicates that the complex reached granulite facies. Even rocks
135 without hyperstene show evidence of high grade metamorphism, such as brown biotite, antiperthitic
136 sodic plagioclase and polygonized textures. Peak temperatures were estimated to 800° (Hartmann
137 1979, 2000) and the pressure to 5–7 kbar (Girardi and Ulbrich 1978; Fornari 1998). This high-grade
138 event was followed by an amphibolite facies retrograde metamorphism and later, greenschist facies
139 mineral assemblages formed along shear zones (Silva 1984; Basei 1985; Basei et al. 1998a). In its
140 southernmost portion, along the Itajaí-Perimbó Shear Zone, highly deformed and reworked portions
141 of the LAT are referred to as São Miguel Complex (Basei 1985). Zircon U-Pb SHRIMP age of tonalite
142 from this complex is 2201 ± 7 Ma (Silva et al., 2000).

143 Available U-Pb, Rb-Sr, Sm-Nd and K-Ar geochronological data (Hartmann et al. 1979, 1998, 2000;
144 Girardi et al. 1974; Basei 1985; Basei et al. 1998a, 1999, 2000, 2009 a,b; Siga Jr 1995; Harara 2001; Sato
145 et al. 2008, Passarelli et al. 2018) indicate an Archean to Paleoproterozoic evolution of the basement
146 of the LAT with tectonic stabilization at the end of the Paleoproterozoic. However, most existing
147 radiometric zircon ages were obtained by TIMS analyses and might partly result from mixed age
148 populations. The oldest zircon U-Pb ages fall in the range of 2.7–2.6 Ga (SHRIMP, Hartmann et al.
149 2000), Rb-Sr and Sm-Nd whole rock analyses yield similar results (Siga Jr 1995). These ages suggest
150 Archean protoliths for the SCGC, even though the rocks were intensely overprinted by high-grade
151 Paleoproterozoic metamorphism. Radiometric ages around 2.35 Ga can be found throughout the
152 SCGC and are attributed to a high grade metamorphic event (Siga Jr 1995; Basei et al. 1998a, 1999,
153 2000, 2009b). U-Pb ages of 2.18 Ga in granulitic paragneisses indicate a second high grade
154 metamorphic event, which apparently took place after a period of erosion and deposition of
155 sedimentary rocks (Basei et al. 1998a, 1999, 2009b). Harara (2001) reports U-Pb ages (TIMS-ID) of
156 2.06 Ga which the author interprets as high grade metamorphic event. Sm-Nd TDM model ages are
157 mainly Neoproterozoic and Siderian in age with individual ages being slightly older or younger (Harara
158 2001; Siga Jr 1995). K-Ar ages which mark the period of tectonic stabilization range from 2.1 to 1.7 Ga
159 with a main peak for biotite at ca. 1.8 Ga (Girardi et al. 1974; Hartmann et al. 1979; Machiavelli 1991;
160 Siga Jr 1995; Harara 2001). Younger K-Ar ages and one monazite U-Pb age of ~ 0.6 Ga were found
161 near shear zones (Basei 1985, Siga Jr 1995).

162 The LAT was affected in the Neoproterozoic by localized granitic magmatism and the development of
163 a set of volcano-sedimentary basins. The Serra do Mar Suite (Kaul 1984), alternatively named
164 Graciosa Province (Gualda and Vlach 2007a,b), consists of A-type alkaline–peralkaline granitoid
165 bodies which stretch parallel to the modern coastline. They are intrusive into the gneissic migmatitic
166 basement and commonly carry xenoliths of gneissic rocks (Basei et al. 1990; Kaul 1997; Siga Jr et al.
167 1997, 1999). Geophysical, structural and textural characteristics as well as the correlation to the
168 volcano–sedimentary basins indicate an emplacement at shallow crustal levels (Hallinan et al. 1993;
169 Gualda and Vlach 2007a,b). The emplacement of the Serra do Mar Suite was at about $580\text{--}583 \pm 3$
170 Ma, extensional A-type magmatism occurred ca. 10–30 Ma later, after the final amalgamation of the
171 LAT, Curitiba and Paranaguá terranes (Vlach et al. 2011).

172 Along the northern portion of the LAT, the Campo Alegre, Corupá and Guaratubinha basins share
173 similar lithological associations, and are therefore considered remnants of the same
174 volcanosedimentary cover (Basei et al. 1998b). They mostly comprise terrigenous sediments covered
175 by thick pyroclastic and volcanic sequences (Citroni et al. 2001; Quiroz-Valle et al. 2019), the onset of
176 deposition of the lowermost unit of the Campo Alegre was constrained to $\sim 606\text{--}590$ Ma (Quiroz-Valle
177 et al. 2019). Together with the A-type magmatism of the Serra do Mar Suite, these basins represent
178 an important extensional phase after in the final stages of the Brasiliano/Pan-African orogenic cycle
179 (Campos Neto and Figueiredo 1995; Campos Neto 2000; Almeida et al. 2010).

180 The other major Neoproterozoic sequence overlying the LAT is the Itajaí Basin (Rostirolla et al. 1999;
181 Guadagnin et al. 2010; Basei et al. 2011b; Costa and Nascimento 2015), which covers much of the
182 southern portion of the terrane. It is an elongated asymmetrical basin, with a total thickness that
183 may add up to 5 km (Basei 1985; Basei et al. 1998b). The sedimentary sequence records basal
184 polymitic conglomerates and massive sandstones, overlain by turbiditic rhythmites and immature
185 sandstones and capped by silty-argillitic and silty-sandy sediments (Rostirolla et al. 1999; Guadagnin
186 et al. 2010; Basei et al. 2011b; Costa and Nascimento 2015). Sedimentation began at ca. 595 Ma and
187 ended by 560-550 Ma with the intrusion of rhyolitic dikes and domes in the upper sequences
188 (Guadagnin et al. 2010; Basei et al. 2011b). New LA-ICP-MS U-Pb zircon data establish a minimum
189 depositional age of 563 ± 3 Ma and the Ediacaran fossil record indicates that the Itajaí Basin contains
190 one of the oldest records of the Ediacaran biota in Gondwana (Becker-Kerber et al. 2020). The basin
191 was affected by two deformation phases, causing local folding and numerous repetitions, and was
192 intruded by the late Subida Granite at ca. 520 Ma (Basei et al. 2008, 2011b).

193 3. Analyzed samples

194 In order to expand our understanding of the evolution of the Paleoproterozoic crust of the LAT, a
195 lithologically complex outcrop, representative of the metamorphic units of the central exposures of
196 the terrane, was chosen for detailed geochronological analyses. The outcrop is a ca. 10 m long and
197 2.5 m high roadcut close to São Bento do Sul (Fig. 1), along the highway that connects it to the town
198 of Corupá (BR-280, coordinates: $26^{\circ}17'57''\text{S}/49^{\circ}23'52''\text{W}$ datum: WGS-84). The predominant
199 lithology of the outcrop is banded gneisses with a strong foliation marked by a 10 to 30 cm thick
200 banding (average attitude $\text{N}090^{\circ}/70^{\circ}\text{S}$) with tight isoclinal folds (Fig. 2A and B). A ca. 25 cm thick
201 almond-shaped amphibolite boudin is hosted in gneiss in the northern portion of the outcrop (Fig.
202 2C), while in the southern portion there is a ca. 35 cm thick pegmatite dyke parallel to the banding
203 (Fig. 2D). Four samples representing all lithologies were selected for U-Pb geochronology and Hf
204 zircon isotopic analysis.

205

206 3.1. Orthogneiss (Samples TRX-04B and TRX-04C)

207 The predominant unit of the outcrop is a relatively homogeneous banded orthogneiss with a well-
208 defined and continuous banding (Fig. 2B). Slight variations in the mineralogical composition occur
209 between different bands, possibly suggesting some degree of magmatic heterogeneity in the
210 protolith. Two samples were selected reflecting this discrete variety. *Sample TRX-04B* (Fig. 3A-B)
211 represents a light grey fine- to medium-grained felsic band with syenogranitic composition. The rock
212 typically has a granoblastic texture in which crystals have polygonal borders indicative of static
213 crystallization. Mafic minerals include amphibole and biotite and are restricted to very thin layers
214 that comprise less than 5% of the rock. Note that, in spite of the expected regional granulite facies
215 context, no pyroxene is present in the sample and the gneiss has an equilibrated amphibolite-facies
216 paragenesis. Accessory minerals include titanite, zircon and opaque minerals, as well as chlorite,
217 epidote and calcite, which are indicative of retrometamorphism. This process is also suggested by the
218 intense alteration of feldspars by means of sericitic and saussuritic transformations.

219 *Sample TRX-04C*, on the other hand, represents a more mafic band of the orthogneiss (Fig. 3C). The
220 rock is a dark grey fine-grained gneiss with quartz-monzonitic composition. Foliation is defined by the
221 alignment of amphiboles and biotites characterizing a nemato-lepidoblastic texture with mostly
222 sutured crystal boundaries. Amphiboles are xeno- to subidioblastic and have strong pleochroism
223 varying from pale yellowish-green to bluish-green. Titanite and zircon are accessory minerals, and

224 later hydrothermal alteration is suggested by the presence of epidote and chlorite, as well as by the
225 alteration of feldspars.

226 3.2. Amphibolite (Sample TRX-04D)

227 The boudin-shaped amphibolite lens in the northern portion of the outcrop is represented by
228 *Sample TRX-04D* (Fig. 3E-G). Foliation in this rock is less defined than in the orthogneiss, and is mostly
229 characterized by the alignment of mafic mineral aggregates, which compose up to 30 % of the rock.
230 The main minerals are plagioclase (ca. 60%) and amphibole (ca. 20%), with subordinate epidote and
231 biotite. Amphiboles in thin section are very similar to those observed in the orthogneiss, and the rock
232 has sutured crystal boundaries. Chlorite, calcite and the intense alteration of plagioclase indicate late
233 retrometamorphism. Considerable amounts of epidote (up to 10%) may also be related to this event,
234 or could be representative of the main metamorphic paragenesis, due to its elevated quantity and
235 apparent equilibrium with biotite.

236 3.3. Pegmatite (Sample TRX-04A)

237 Lastly, *Sample TRX-04A* (Figs. 2D and 3H) represents the pegmatitic dyke in the southern portion of
238 the outcrop. The dyke is parallel to the banding and foliation of the outcrop and the contact shows
239 some softening of the surrounding orthogneiss, but it has no visible foliation and has an igneous
240 texture. The pegmatite is salmon-colored and is dominated by very coarse grained feldspar crystals
241 of several centimeters. As with the remaining rocks of the outcrop, the pegmatite was affected by
242 later hydrothermalism, as indicated by intense sericitic and saussuritic transformation in feldspar and
243 epidote-filled veins.

244 4. Results and mineralogical discussion

245 4.1. U-Pb geochronology

246 All U-Pb measurements were performed using the LA-MC-ICP-MS technique, following procedures
247 detailed in Electronic Supplement S1. Concordia plots and weighted average ages calculated during
248 interpretation of the dataset were performed using Isoplot/Ex 3.7 (Ludwig 2001). In total, 199 spot
249 analyses were performed in zircon, however, 57 of them needed to be rejected due to high common
250 lead contents or unrealistic concentrations after data reduction. In titanite, 65 spot analyses were
251 performed yielding quite well constrained results and only two analyses needed to be rejected due
252 to the same reasons. The high number of analyses with analytical difficulties and the difference
253 observed in the overall consistency of the U-Pb dataset for titanite and zircon are probably a result of
254 intensive hydrothermal alteration affecting both minerals differently. Spots that yielded results with
255 analytical problems were discarded and were not considered during the interpretation of the U-Pb
256 dataset. All remaining analyses, including both concordant and discordant results, are presented in
257 the Electronic Supplements S2 and S3 organized according to the interpretation of each spot analysis.
258 Because most of the concordant results yield Paleoproterozoic to Archean ages, whenever individual
259 ages are mentioned they correspond to $^{207}\text{Pb}/^{206}\text{Pb}$ ages, except if noted otherwise.

260 4.1.1. Titanite results

261 Titanite U-Pb results are discussed first in this section because they are much more uniform than
262 those of zircon, providing a rather straightforward interpretation that can be used as a geological
263 constraint for interpreting the rest of the dataset. All results can be found in the Electronic
264 Supplement S2.

265 A total of 33 spot analyses were performed in crystal fragments of sample *TRX-04C* (mafic
266 orthogneiss). Analyzed crystals have common Pb contents mostly between 4 and 20%, and isotopic
267 results are uniform with concordant single-spot ages between 2.05 and 1.96 Ga. A mean $^{207}\text{Pb}/^{206}\text{Pb}$
268 age of 1.991 ± 0.021 Ga was calculated considering all 33 spots (Fig. 4).

269 Results from sample *TRX-04D* (amphibolite) also have common Pb values up to 20%, but yielded
270 more scattered isotopic ratios. A mean $^{207}\text{Pb}/^{206}\text{Pb}$ age of 2.027 ± 0.028 Ga was calculated using 28 of
271 the 30 measured spots rejecting two outlying analyses (Fig. 4).

272 Both samples produced quite similar ages that are overlapping within uncertainty, resulting in a
273 confident constraint of the timing of titanite crystallization in the studied lithological association at
274 ca. 2.02-1.99 Ga. As will be discussed in Section 4.1.2., this particular time interval is notably absent
275 in the dated zircon crystals from all rocks that have a penetrative metamorphic foliation. This suggest
276 that the crystallization event recorded in the titanites took place under metamorphic conditions in
277 which no new zircon crystals were crystallized and that promoted no overgrowth of metamorphic
278 zircon. Hence, this event could be associated with the amphibolite-facies re-equilibration observed in
279 all analyzed metamorphic rocks, presumably responsible for the substitution of the granulite-facies
280 orthopyroxenes described in most occurrences of the Santa Catarina Granulitic Complex. This is in
281 accordance with the fact that titanite in metabasic rocks is more stable in amphibolite facies than in
282 granulite facies, where it is usually substituted by rutile or ilmenite (Frost et al. 2001; Kohn 2017).
283 This interpretation is coherent with the geochronological evolution suggested for the rest of the
284 dataset, but challenges previous mineralogical and geochemical observations that assumed a
285 magmatic origin for titanite in syenitic rocks of the LAT (Hartmann et al. 1998).

286 4.1.2. Zircon results

287 Zircon crystals from all four samples show many different textures in cathodoluminescence imaging,
288 some of which are rather complex and chaotic (Fig. 5A). Several grains contain textures typical for
289 high grade metamorphism such as irregular concentric zoning, recrystallization in parts of the crystal
290 or flow structures (Corfu et al. 2003; Taylor et al. 2016), examples are presented in Fig. 5A. All four
291 analyzed samples show a similar transformation pattern which is exposed in the CL-images. The
292 transformation is visible in form of so called spongy or porous textures (for simplicity only referred to
293 as spongy in the following) (Fig. 5A). The samples are affected to different degrees, with the
294 strongest modification seen in the pegmatite, which is the youngest sample of the outcrop and
295 shows the most idiomorphic grains. In Corfu et al. (2003) and Nasdala et al. (2010) such spongy
296 textures are described as a result of hydrothermal alteration. A more detailed discussion of this topic
297 follows further down.

298 The transformation complicates the U–Pb analysis and the data interpretation. The loss of previous
299 textural information inhibits a discrimination of the different parts of the zircons, and hydrothermal
300 processes may lead to heterogeneous chemical distributions in crystals (e.g. Geisler et al. 2003;
301 Anderson et al. 2008). In addition to the textural problems, the zircons of all four samples have many
302 fractures and the possibilities of good spot locations are limited. In contrast to the titanite results,
303 the U-Pb dataset obtained from zircon is characterized by a high number of discordant analyses with
304 more than one third of the analyses being >10% discordant, many of which exhibit reverse
305 discordance (Figs. 6 and 7). All results can be found in the electronic Supplement S3. Relating the
306 quality of the analyses (including the rejected spots) to the textures (see Fig. 6) shows that the
307 proportion of problematic and highly discordant analyses is highest in the spongy grains and those
308 showing textures classified as “broad altered zoning”, characterized by remnants of broad zoning,
309 often core zoning, which shows alteration at the contact of the different zones (for examples see Fig.

310 5A). On the other hand, best results were obtained in homogeneous bright portions of the grains
311 (Fig. 6).

312 4.1.2.1. Causes for discordance and dispersion

313 The zircon dataset shows a remarkable dispersion and even using traditional discordance filters, such
314 as, for example, considering analyses that are no more than 10% discordant, the combined dataset
315 for all samples is spread over an interval of over 1.5 Gyr (3.20 – 1.58 Ga) (Fig. 7A and B). U-Pb dates
316 of granulitic rocks often disclose significant age dispersions and the capability to discriminate
317 between protracted crystallization, age bias due to radiation damage induced Pb-loss and analytical
318 uncertainty is expected. Our data does not describe neither a normal distribution of concordant
319 analyses along the Concordia curve nor a single Discordia trend with well-constrained intercepts (see
320 Fig. 7A). This suggests that multiple populations with different ages were affected by an opening of
321 the isotopic system, resulting in a disposition along the Concordia curve and multiple Discordia
322 trends. A partial resetting of older zircon crystals could have been caused by the amphibolite-facies
323 event recorded in the crystallization of titanite at ca. 2.01 Ga. The crystallization of titanite
324 establishes a geological constraint for the retrometamorphism post-dating the thermal peak of the
325 rocks that have a penetrative metamorphic foliation, that is, the orthogneisses and the amphibolite
326 *boudin* represented by samples TRX-04B, C, and D. This event predates the intrusion of the later
327 pegmatite dyke represented by sample TRX-04A, as evidenced by its lack of a discernible
328 metamorphic foliation and characteristic igneous texture. This is reflected in the combined zircon U-
329 Pb dataset, which can roughly be divided into two distinct groups (Fig. 7A). Results from samples of
330 the metamorphic rocks are characterized by ages predominantly older than 2.1 Ga, with younger
331 ages recorded only in the more discordant analyses. On the other hand, zircons from the pegmatite
332 dyke recorded only a few crystals older than 2.0 Ga, and no results older than 2.3 Ga. These
333 observations confirm the interpretation that the emplacement of the pegmatite dyke took place
334 after the amphibolite-facies re-equilibration recorded by titanite in the orthogneiss and amphibolite.
335 Nonetheless, many zircons of sample TRX-04A show alteration textures and many spot analyses are
336 highly discordant or even needed to be rejected. This implies that at least one event causing
337 widespread partial Pb loss in the U-Pb dataset is not contemporaneous to the crystallization of
338 titanite, and instead post-dates the emplacement of the pegmatite dyke.

339 The actual age of this event is likely much younger, based on the position close to Neoproterozoic
340 ages of the most discordant crystals along the Concordia curve (Fig. 7A). Because the LAT does not
341 record regional thermal overprint during the Neoproterozoic Brasiliano orogenic cycle, as indicated
342 by Paleoproterozoic K-Ar cooling ages (Siga Jr. 1995), this Pb loss is interpreted to have happened
343 under low-temperature conditions during a hydrothermal event. As zircon is usually very resistant to
344 hydrothermal alteration, the susceptibility of zircon crystals to hydrothermal fluids in low-
345 temperature conditions is strongly controlled by the integrity of the crystalline structure of the
346 mineral (e.g. Rubatto 2017 and references therein). Spontaneous decay of radioactive elements
347 (mainly U and Th) leads to damage of the crystalline structure as alpha-recoil produces small
348 amorphous domains (Nasdala et al. 2001; Palenik et al. 2003). This damage accumulates if the
349 temperature does not allow natural annealing of structure, leading to the progressive
350 metamictization of the crystal. Leaching experiments in metamict zircon show that metamictization
351 produces a significant decrease in stability against hydrothermal fluids (Geisler et al. 2001). Although
352 the zircons analyzed here, including the ones of the pegmatite, have generally low or normal U-
353 contents, the enormous time interval between the magmatic and metamorphic events recorded in
354 the samples (>1.8 Ga) and the assumed hydrothermal event (ca. 600-550 Ma) provides enough time
355 for the accumulation of sufficient radiation damage for the (partial) metamictization of zircon. Hoskin

356 (2005) estimates that, for U and Th compositions typical of magmatic zircons, time intervals in the
357 order of a few hundreds of Myr would be sufficient to enable hydrothermal Pb disturbance.

358 Although Pb diffusion can be also very significant in titanite and is affected by radiation damage
359 (Cherniak 1993), this mineral is usually less affected by partial Pb loss and yields therefore often
360 more concordant U-Pb ages than zircon (Tilton and Grünenfelder 1968). This is probably due to
361 differences in annealing and diffusion behavior and typically lower concentrations of U and Th
362 (Howie et al. 1992) and thus lower doses of radiation damage. This may explain the contrast between
363 very concordant titanite and the rather disturbed zircon results observed in our dataset.

364 Figure 8 displays the relationship between content of radioactive elements (a proxy for the likely
365 intensity of radiation damage, described in terms of effective Uranium: $eU = U + 0.235 \text{ Th}$ in ppm), U-
366 Pb age, concordance and textures. The figure evidences that different textures predominate in
367 different samples and different age groups. Most young, highly discordant analyses belong to grains
368 with alteration textures (spongy and “broad altered zoning”), particularly in sample TRX-04A. A
369 detailed look on this relation is given by the comparison between the actinide content of each
370 crystal, expressed by its eU which is a proxy for the radiation damage, and the degree of discordance
371 (Fig. 8B) and resulting ages (Fig. 8C, expressed in the more susceptible $^{238}\text{U}/^{206}\text{Pb}$ system). For the
372 normal discordant U-Pb data (discordance < 100%), mainly belonging to spots with alteration
373 textures, eU correlates strongly with the degree of concordance (Fig. 8B). Similarly, the younger U-Pb
374 ages correlate with eU (Fig. 8C). The correlation is clearer when comparing eU to the degree of
375 concordance than to the overall ages, as the altered grains belong to different age populations.
376 These observations point to the hydrothermal nature of the younger (Neoproterozoic) opening of the
377 U-Pb isotopic system and to radiation damage as main aspect controlling the Pb loss. The
378 correlations are most evident in sample TRX-04A as this sample comprises the highest number of
379 highly discordant analyses and spots on altered textures (see Fig. 8) and furthermore has a more
380 homogeneous age distribution (see below). This sample is therefore the best candidate for an
381 estimate of the timing of the Pb loss by the calculation of a Discordia trend, as will be attempted in
382 Section 4.1.2.2. Note that alteration is also present in the metamorphic samples (TRX-04B/C/D),
383 however less pervasive, so that unaltered spot locations were easier to find, as reflected in their
384 relatively less disturbed dataset.

385 Because of the probable opening of the U-Pb isotopic system identified in the dataset, additional
386 precautions have to be taken for its interpretation and, particularly, for the calculation of geologically
387 meaningful ages. As discussed by Spencer et al. (2016), the application of traditional discordance
388 filters, such as the acceptance of all analyses $\leq 10\%$ discordant, for datasets which underwent
389 significant Pb loss can lead to misleading interpretations, especially in ancient samples. One strategy
390 suggested by these authors for dealing with such datasets is to consider only analyses for which the
391 results intersect the Concordia curve within analytical uncertainties. In our dataset, this approach
392 leads to the acceptance of only 35 crystals out of a total of 142 analyses, in a distinct contrast to the
393 population of 92 spot analyses that have ages $\leq 10\%$ discordant (Fig. 7B). However, comparing the
394 distribution of $^{207}\text{Pb}/^{206}\text{Pb}$ ages for groups of analyses with different concordance values shows
395 remarkably similar results, even when considering the entire dataset, that is, including strongly
396 discordant analyses (Fig. 7B). The main differences between the different distribution curves are that
397 the main population peaks tend to broaden when including less concordant results, together with a
398 progressive increase in the youngest populations, creating a longer tail that skewers the results
399 towards younger results. Both of these processes are reflective of how small increases in the degree
400 of discordance lead to a higher dispersion of the U-Pb dataset. Nonetheless, the position of the main
401 peaks and their relative prominence remains similar irrespective of the adopted criteria. This shows
402 that the highly concordant analyses, which will be used for refined geological interpretations and age

403 calculations as they are least affected by Pb loss, are representative of the geochronological
404 signature of the entire dataset in spite of their reduced number. Furthermore, it demonstrates that
405 the adoption of slightly more discordant $^{207}\text{Pb}/^{206}\text{Pb}$ ages (e.g. up to 10% discordance) is acceptable
406 for subsequent analyses when the consideration of a larger dataset is necessary, such as for the
407 calculation of Hf-based model ages (Section 4.2), as they reproduce the overall distribution of the
408 more precise analyses.

409

410 4.1.2.2. Zircon crystallization ages for sample TRX-04A

411 Only 6 out of 47 analysed zircon crystals of sample A have results that intersect the Concordia curve
412 within analytical uncertainty (Fig. 9A). Two spot analyses for rounded /ovoid homogeneous dark
413 crystals give Rhyacian ages (2.29 and 2.16 Ga) that overlap with ages of the high-temperature
414 metamorphic events recorded in the remaining samples (Section 4.1.2.3.) and precede the
415 amphibolite-facies event recorded by the crystallization of titanite. As discussed above, this
416 metamorphic event precedes the emplacement of the pegmatite dyke, and therefore these crystals
417 are considered inherited. The other analyses correspond to two pairs of crystals with Orosirian to
418 Statherian ages at ca. 1.96 Ga and 1.79 Ga that are contemporaneous and younger than the last
419 amphibolite-facies metamorphic event (for exemplary grains and ages see Fig. 5B).

420 Either of these two sets of ages could represent the actual magmatic age of the dyke, with somewhat
421 different consequences for the geological evolution of the area. If the younger population (1.79 Ga)
422 corresponds to the emplacement, the population at ca. 1.96 would correspond to inherited
423 xenocrysts from a time period that is conspicuously absent from the remaining samples (Fig. 7A and
424 9A-D). In this sense, if the emplacement of the dyke happened at ca. 1.79 Ga, the crystals at ca. 1.96
425 Ga could either represent xenocrysts from a restricted magmatism, possibly associated with the
426 amphibolite-facies event at ca. 2.01 Ga, or mixed ages between inherited cores and overgrown rims.
427 Unfortunately, as the grains are either homogeneous dark or spongy, initial textures cannot be
428 detected. On the other hand, if the older population (1.96 Ga) represents the emplacement age, than
429 the younger age should represent magmatic crystals affected by partial Pb loss.

430 In addition, as discussed in Section 4.1.2.1., sample A is the best candidate to estimate an age for the
431 Neoproterozoic Pb loss. A calculated Discordia trend for sample A (not displayed) has an upper
432 intercept with an age of $2,063 \pm 81$ Ma and a lower intercept of 515 ± 150 Ma. The remarkably large
433 uncertainties in these intercept ages are probably caused by the fact that there is more than one
434 population of Paleoproterozoic concordant crystals recognized in the sample, preventing the
435 distribution of the data along a single Discordia line. Furthermore, the zircon grains may have
436 suffered some recent Pb loss due to their current exposure to tropical weathering. Nonetheless, the
437 lower value of 515 ± 150 Ma calculated for sample TRX-04A is geologically sensible, as the main
438 geological events that affect the LAT, such as granitic magmatism, shear zone reactivation,
439 development of a volcanic basin, and associated hydrothermalism, all took place during the
440 Brasiliano orogenic cycle between 600 and 560 Ma. The inset in Figure 7C shows how a
441 Neoproterozoic Pb loss event could lead to an age distribution similar to the one observed in sample
442 04-A.

443 4.1.2.3. Zircon crystallization ages for samples TRX-04B/C/D

444 Samples TRX-04B, C and D represent associated lithologies that, in the field and in thin section, are
445 interpreted to have experienced a common geological history during deformation and regional
446 metamorphism. Accordingly, they share complementary U-Pb results, and will be treated together.
447 Out of the 110 spot analyses successfully performed for the three samples, a total of 29 crystals

448 produced results that intercept the Concordia curve within analytical uncertainty (TRX-04B: 12; TRX-
449 04C: 7; TRX-04D: 10). While the different samples record populations that have overlapping ages (Fig.
450 9B-D), each of them has no more than a few analyses in a given sample. Therefore, in order to
451 calculate more precise and statistically robust ages representative of the entire dataset, results from
452 different samples but representative of a same population are considered together.

453 In total, 5 populations are recorded in the three samples, exemplary grains of each population are
454 presented in Figure 5B. The two youngest ones are recorded in the majority of crystals, being
455 identified in all samples. The largest of these is a group of 13 spots with individual ages between 2.21
456 and 2.12 Ga that, together, have a weighted mean age of 2.183 ± 0.017 Ga. The other population
457 consists of 6 crystals with ages from 2.36 to 2.29 Ga, for which a Concordia age was calculated at
458 2.352 ± 0.017 Ga. Because these populations are shared by all samples and represent the youngest
459 concordant crystals recorded in them, they probably represent metamorphic crystallization events,
460 possibly associated with the regional granulite-facies event identified in the LAT.

461 The remaining age populations are more elusive and have a less systematic record in the different
462 samples, ultimately forming a less constrained picture. Ages at ca. 2.5 Ga were recorded only in two
463 crystals in sample TRX-04B, producing a Concordia age of 2.498 ± 0.011 Ga. A somewhat more
464 numerous yet disperse population was identified in five crystals samples TRX-04B and D with
465 individual ages between 2.75 and 2.60 Ga. The best multi-crystal estimate for this population is a
466 rather imprecise weighted mean of 2.683 ± 0.017 Ga. Finally, two inherited cores from sample TRX-
467 04B record stand-alone Meso-to Paleoproterozoic ages of ca. 3.20 and 3.09 Ga. All of these crystals are
468 interpreted to record the magmatic processes responsible for the generation of the protoliths of the
469 metamorphic samples. The more numerous population probably records the main period of
470 magmatic activity, which already included some degree of recycled crust, as indicated by the
471 presence of two inherited crystals with ages older than 3.0 Ga.

472 Differently from sample TRX-04A, discordant analyses in samples B, C and D are predominantly
473 *reverse* discordant analyses (63 out of 81 discordant analyses), with few *normal* discordant analyses
474 (18 out of 81 discordant analyses). The *normal* discordant analyses fall into the area between the
475 oldest concordant ages, the titanite crystallization and the Neoproterozoic-Cambrian Brasiliano-
476 event (Fig. 7C). The complexity created by the multiple populations and at least one, possibly more
477 Pb-loss events does not allow the calculation of a single Discordia trendline.

478

479 4.1.3. Reverse Discordance

480 Natural reverse discordance is not very common in zircon U-Pb datasets and mainly reported from
481 Archean rocks (Williams et al. 1984; Corfu 2013; Kusiak et al. 2013; Wiemer et al. 2017). In most of
482 the cases reverse discordance seems to be related to high-temperature metamorphism, but Wiemer
483 et al. (2017) present an example where reverse discordance was apparently evoked by a Paleozoic Pb
484 loss event under low T conditions. While the exact mechanism and reasons for reverse discordance
485 remain a matter of debate in the afore mentioned references, it seems that reverse discordance is
486 linked to nano and micro-scale redistribution of radiogenic Pb. Kusiak et al. (2015) observed pure
487 metallic Pb inclusions in zircon which are probably the most extreme case of Pb redistribution.
488 Utsonomiya et al. (2004) report that Pb diffusivity is enhanced when the crystal lattice is already
489 radiation damaged. Wiemer et al. (2017) observe reverse discordance mainly in low eU areas of
490 grains and explain that reverse discordance is produced by migration of radiogenic Pb from high eU
491 areas to low eU areas during a Pb loss -or rather Pb redistribution- event under low T conditions.

492 In our dataset, reverse discordance occurs mainly in grains / grain areas with homogeneous dark
 493 texture (see Fig. 8). There is a clear relationship of reverse discordance with eU with reverse
 494 discordance being strongest in very low eU spots (see Fig. 8). This supports the hypothesis that
 495 reverse discordance is produced by implantation of radiogenic Pb, as these spots are the most
 496 sensitive to the addition of radiogenic Pb, and even the addition of small amounts of radiogenic Pb
 497 can have a strong impact on their U/Pb ratios.

498 Figure 7C indicates how multiple Paleoproterozoic age populations affected by an Neoproterozoic Pb
 499 redistribution event could produce a data distribution similar to the one observed in our data set.
 500 This supports the idea that in our case, as for Wiemer et al. (2017), reverse discordance was evoked
 501 by the same event which caused Pb loss. This event not only caused the damage of the zircon
 502 crystalline lattice and the loss of radiogenic material, but caused chemical redistribution inside the
 503 crystals, transporting radiogenic Pb from areas rich in U and Th (and thus radiogenic Pb) into areas
 504 poor in U, Th and radiogenic Pb, causing reverse discordance in the latter. Preexisting radiation
 505 damage of the crystalline structure, accumulated during interval of >1.2 Gyr between zircon
 506 crystallization (>1.8 Ga) and the hydrothermal event (600-550 Ma), probably facilitated this process
 507 by increasing Pb mobility. Although it seems likely that the Neoproterozoic hydrothermal event was
 508 the factor leading to reverse discordance, we cannot exclude that the previous high temperature
 509 metamorphism of the rocks played a role, too. More research will be needed in order to better
 510 understand the involved processes and their effects.

511 4.2. Hf isotopic data

512 Previously dated zircon crystals with the highest degree of concordance of the most frequent age
 513 populations were chosen for the measurement and 69 hafnium spot analyses were performed on the
 514 same spots as U-Pb analyses. All results can be found in the Electronic Supplement S4. In contrast to
 515 the U-Pb analyses, no analytical difficulties were identified complicating the Hf analyses, resulting in
 516 overall reliable results. The undisturbed Hf results are in line with observations by Gerdes and Zeh
 517 (2009) and Lenting et al. (2010) indicating that Hf isotopes remain nearly unaffected during alteration
 518 processes which produce strong Pb loss and disturbed U-Pb ages in zircon.

519 All but three analyses yield negative $\epsilon_{\text{Hf}}(t_1)$ values as low as -16.8, and Hf T_{DM} model ages cover a
 520 continuous range from 4.3 to 2.7 Ga (Fig. 11A and B). The detailed description of the results will be
 521 performed in the same sample groups as for the U-Pb ages.

522

523 4.2.1. Sample A

524 The 10 Hf spot analyses performed on sample A yield $\epsilon_{\text{Hf}}(t_1)$ from -4.2 to -13.6 and T_{DM} model ages
 525 from 3.2 - 2.8 Ga (see Fig. 10A). The Hf T_{DM} model ages are rather homogeneous and show no
 526 systematic distribution or interdependencies. Reducing the dataset to those analyses in which the U-
 527 Pb ellipses intersect the Concordia curve does not change the general distribution of the dataset. The
 528 concordant analyses at ca. 1.8 and 2.0 Ga considered to date the emplacement of the pegmatite
 529 yield Hf T_{DM} model ages of 3.1-2.8 Ma indicating a long crustal residence.

530 4.2.2. Samples B/C/D

531 The $\epsilon_{\text{Hf}}(t_1)$ values of samples of metamorphic rocks range from 1.3 to -16.8, with Hf T_{DM} model ages
 532 between 4.3 and 2.7 Ga (Fig. 10A). The spot analyses form 2-3 clusters when comparing U-Pb age
 533 with $\epsilon_{\text{Hf}}(t_1)$. As for sample A, considering only those analyses for which U-Pb ellipses intersect the
 534 Concordia does not change the general distribution of the Hf results.

535 The main cluster is composed of crystals with U-Pb of 2.5-2.0 Ga, $\epsilon_{\text{Hf}}(t_1)$ values of 1.3 to -11.4 and T_{DM}
 536 model ages of 2.7-3.37 Ga. The second cluster is formed by analyses with U-Pb ages > 2.365 and T_{DM}
 537 model ages > 3.47 Ga. The oldest inherited cores with U-Pb ages >3 Ga may form a distinct
 538 population, but due to the limited amount of data it is not possible to confidently distinguish it from
 539 the latter cluster.

540 Although there is an overall negative correlation of U-Pb ages and $\epsilon_{\text{Hf}}(t_1)$, the two main clusters share
 541 a similar pattern marked by a positive negative correlation between U-Pb age and $\epsilon_{\text{Hf}}(t_1)$ (see Fig.
 542 10A and B). This suggests two pulses of differentiation and crystallization, each of them starting with
 543 the crystallization of more juvenile material followed by an increase in the participation of recycled
 544 material. In both clusters, the material with the most crustal signatures (i.e. more recycled material),
 545 is added towards the end of each cycle. On the other hand, crystals with the most juvenile signature,
 546 characterized by positive $\epsilon_{\text{Hf}}(t_1)$ values, mark the beginning of the youngest cycle.

547 The zircon crystallization events at ca. 2.5 Ga and 2.35 Ga (see Fig. 10B) are noteworthy in that they
 548 contain Hf signatures from both clusters, indicating that in these events preexisting material was
 549 mixed with new and more juvenile material. On the other hand, the event at ca. 2.2 Ga apparently
 550 involved a complete mixture of the two endmember compositions: the strongly crustal, recycled
 551 material with old model ages (>3.47 Ga) and lag times, as well as the juvenile material with positive
 552 $\epsilon_{\text{Hf}}(t_1)$ which appears the first time at 2.5 Ga.

553 5. Regional Implications

554 5.1. Archean and Paleoproterozoic evolution of the LAT

555 The data presented in this contribution reflect the long and complex evolution of the LAT, as
 556 suggested previously by other authors (e.g. Basei et al. 1998a; 2009b; Passarelli et al. 2018). In
 557 particular, the wide spread of geologically meaningful crystallization ages even in individual samples
 558 illustrates the importance of applying high-resolution geochronological analyses in this type of rock.
 559 As such, the new data represents a complement to the available data which, except for a handful of
 560 SHRIMP analyses, were predominantly obtained by ID-TIMS (Hartmann 2000; Basei 2009 a, b; Sato et
 561 al. 2008). The possibility that some of these analyses performed with traditional methods represent
 562 mixed ages is probably responsible for most previous Discordia ages.

563 Our data confirms that the LAT has a significant ancient signature. Archean ages had been previously
 564 obtained with Rb-Sr WR analyses and imprecise upper intercept ages from ID-TIMS Discordias (Siga Jr
 565 1995; Hartmann 1979; Basei 1985; Basei et al. 2009a,b), but high-resolution Archean records were
 566 registered in a few SHRIMP U-Pb analyses as old as 2.72 Ga (Hartmann et al. 2000). Our data set
 567 includes concordant Mesoarchean ages of 3.0-3.2 Ga and, more significantly, Hf isotopes indicating
 568 recycled crustal signatures and differentiation from the mantle in the Hadean or Early Archean.
 569 These new Hf T_{DM} model ages are significantly older than existing Sm-Nd T_{DM} model ages, which
 570 range from 3.4 to 2.2 Ga (Siga Jr. 1995; Harara 2001; Basei 2009b; Passarelli 2018).

571 While the oldest ages are restricted to a few zircon xenocrysts, the oldest significant group of ages
 572 yields a mean $^{206}\text{Pb}/^{207}\text{Pb}$ age of $2,683 \pm 70$ Ma. While Hartmann et al. (2000) argue for two events
 573 during the Neoproterozoic, with magmatism at 2.72 Ga followed by high grade metamorphism at 2.68
 574 Ma, our data does not recognize two distinct populations, instead suggesting a period of prolonged
 575 magmatic activity, which agrees with the poly-magmatic evolutions suggested by other authors for
 576 different associations of the LAT (e.g. Basei et al. 2009). . Nonetheless, all Archean crystals in our
 577 dataset come from samples that experienced an important metamorphic overprint in the
 578 Paleoproterozoic and, so far, no undisturbed Archean rocks have been identified in the LAT.

579 Two concordant analyses seem to record an event for which a Concordia age of $2,498 \pm 11$ Ma was
 580 calculated. Although this age population is small, it stands out for its positive $\epsilon_{\text{Hf}}(t_1)$ values (1.3 – 0.6),
 581 indicating that this event is significant for the addition of juvenile material. The igneous textures of
 582 these grains (oscillatory zoning) and the juvenile Hf signature allow a more precise interpretation and
 583 indicate a magmatic origin. Although this age population has only a limited representation in the U-
 584 Pb record, the Hf signature of these analyses and of the grains crystallized afterwards (populations of
 585 2.35 and 2.2 Ga) suggest that newly differentiated material was added at this time. As mentioned in
 586 Section 4.2.1 the zircons of the later events have a Hf signature which seems to be a mixture of the
 587 one of the Archean zircons and of the 2.5 Ga population.

588 The next well-defined zircon population produces a Concordia age of $2,352 \pm 23$ Ma. Ages around
 589 2350 Ma can be found throughout the Santa Catarina Granulitic Complex and are assigned to a high-
 590 grade metamorphic event (Siga Jr 1995; Basei et al. 1998a, 1999, 2000, 2009b). Less than 200 Ma
 591 later, around 2180 Ma the Complex was again subject to high grade metamorphism, as suggested
 592 previously in the literature (Basei 1985; Siga Jr. 1995; Hartmann et al. 2000; Harara 2001; Sato et al.
 593 2008; Basei et al. 2009a, b). The mean $^{206}\text{Pb}/^{207}\text{Pb}$ age of $2,183 \pm 17$ Ma, calculated in this study from
 594 13 spots that intersect the Concordia curve, confirm this interpretation and provide a much more
 595 robust age constraint for the event. Individual crystal ages of this population have a rather
 596 widespread range (2.21 to 2.12 Ga), preventing the calculation of a Concordia age, and may thus
 597 reflect a long-lasting geological event. As the most significant population in our dataset, this event
 598 was probably responsible for the present configuration of the LAT as a high-grade metamorphic
 599 complex. Together with an abundant Paleoproterozoic record in the region, it may record a long-
 600 lived succession of orogenic cycles responsible for the amalgamation of basement associations in
 601 southern South America. The Hf T_{DM} model ages of the two latter high-grade metamorphic events
 602 range from 2.7 Ga to 3.4 Ga and are, as explained before, probably a mixture of two end-member
 603 compositions. They overlap partially with some Sm-Nd T_{DM} model ages presented in the literature
 604 (Siga Jr. 1995; Harara 2001; Passarelli 2018)

605 As mentioned in Section 4.1.1, the titanite mean $^{206}\text{Pb}/^{207}\text{Pb}$ ages of $1,991 \pm 21$ Ma and $2,027 \pm 28$ Ma
 606 are interpreted to date the retrograde metamorphism following the high-grade metamorphic peak in
 607 the LAT that was responsible for the complete re-equilibration of our samples into an amphibolite-
 608 facies paragenesis. Girardi et al. (1974), Basei (1985), Machiavelli et al. (1993), Siga Jr (1995) and
 609 Harara (2001) report Pb-Pb and Rb-Sr WR ages of ~ 2.0 in rocks with little orthopyroxene and
 610 conspicuous presence of amphibole as the main mafic mineral, reinforcing this interpretation. Most
 611 of these ages have significant analytical errors, and therefore our new titanite U-Pb spot data allows
 612 a tighter age constraint of this event between 2.02 and 1.99 Ga.

613 The emplacement of the pegmatite dyke dated in this study, which took place between 1.96 and 1.79
 614 Ga, goes either along with the end of the retrograde metamorphism or falls in the period of cratonic
 615 stabilization described for the LAT. This period is defined by K-Ar cooling ages from hornblende and
 616 biotite extracted from multiple expositions of the granulitic complex with ages of 2.1-1.7 Ga and a
 617 major concentration around 1.8 Ga for biotite (Girardi et al. 1974; Hartmann et al. 1979; Basei 1985;
 618 Machiavelli 1991; Siga Jr 1995; Harara 2001). Concordant Zircon ages of 2.0 or 1.8 Ga are not
 619 described in the literature, nor do they appear in the metamorphic samples studied here, indicating
 620 that the pegmatite was probably formed by a local magmatic event in the late stages of cratonic
 621 stabilization.

622 5.2. The LAT in the context of the Brasiliano/Pan-African orogenic cycle

623 The emplacement of the pegmatite dyke constitutes the last significant geological event in the LAT
 624 prior to the Neoproterozoic Pan-African/Brasiliano orogenic cycle. The fact that undisturbed

625 Paleoproterozoic K-Ar ages are recorded even for minerals with low closure temperature such as
626 biotite (*ca.* 310 °C, Harrison et al. 1985) illustrate how the LAT remained remarkably unaffected even
627 during the intense orogenic processes leading to the assembly of Gondwana in the Late
628 Neoproterozoic.

629 Geological evidences of this event in the LAT affect predominantly its borders. The southern
630 extremity of the terrane is covered by the 650 to 550 Ma old Itajaí Basin (Basei et al. 2010;
631 Guadagnin et al. 2010). The northern portion of the LAT, on the other hand, is patchily intruded by A-
632 type granitoids from the Serra do Mar Suite, which were emplaced at a shallow crustal level between
633 590 and 570 Ma (Basei et al. 2009; Vlach et al. 2011). In the same region, the Campo Alegre, Corupá
634 and Guaratubinha basins are characterized by a predominance of volcanic and pyroclastic deposits
635 overlying continental sediments. The biggest of these sequences, the Campo Alegre Basin, was
636 deposited between *ca.* 605 and 585 Ma (Citroni et al. 2001; Quiroz-Valle et al. 2019; Lino et al. in
637 review).

638 Reflecting this regional pattern, while most of the basement of the LAT records its original subvertical
639 NW-striking structural configuration, both its northern and southern borders have NE-trending
640 structures evidencing a Neoproterozoic overprint (Basei 1985; Harara 2001). In the central portion of
641 the terrane, only localized NE-striking shear zones cross-cut the Santa Catarina Granulitic Complex.
642 These structures are associated with a subordinate population of partially or completely reset K-Ar
643 ages in amphibole and mica, recording an overprint of the geochronological system at *ca.* 660-620
644 Ma (Basei et al. 1998, 2009; Passarelli et al. 2018 and references therein). This is mostly attributed to
645 the percolation of hydrothermal fluids along the fault zones, which also controlled the deposition
646 and widespread metasomatic transformations in the Campo Alegre Basin (Lino et al. in review). The
647 same authors propose an age of *ca.* 565 Ma for the hydrothermal alteration of pyroclastic rocks in
648 this basin, which is in accordance with our interpretation that much of the perturbation of the U-Pb
649 of the present dataset is a possible effect of Neoproterozoic fluid percolation..

650 The tectonic stability since the Paleoproterozoic, noticeable for a relatively small crustal fragment
651 surrounded by two orogenic belts with extensions of hundreds to thousands of kilometers, has led to
652 the classification of the LAT as a microplate or microcraton during the assembly of West Gondwana
653 (e.g. Basei et al. 1998, 2009, 2010). The lack of a significant Neoproterozoic overprint, in contrast
654 with the highly reworked basement inliers immediately to the north (Curitiba Terrane) was
655 interpreted as an indication that the LAT constitutes an exotic terrane bound by suture zones. The
656 most commonly proposed configuration of this model interprets the LAT and Curitiba Terrane to
657 have been juxtaposed during the Brasiliano Orogenic Cycle along a former subduction zone
658 represented by the Piên Shear Zone (Basei et al. 2000, 2008, 2009, 2018; Passarelli et al. 2018). In
659 this model, the subduction would be responsible for the generation of a magmatic Arc between 615
660 and 595 Ma (Piên Suite) and the obduction of remnant oceanic crust (Piên mafic-ultramafic Suite)
661 (Harara 2001).

662 Alternatively, the lack of remnants of passive margin sediments between the LAT and the Curitiba
663 Terrane, together with similarities in the isotopic signatures and Archean to Paleoproterozoic
664 evolution, have been used to propose a common origin for the two terranes, in which their main
665 difference would be the contrasting degrees of tectonic reworking during the Brasiliano Orogenic
666 Cycle (Siga Jr et al. 1993). This model has similarities with recent proposals that suggest a common
667 origin not only for these two terranes, but also for additional basement remnants in southern Brazil
668 and Uruguay, such as the Encantadas and Nico Pérez Terranes (Oyhantçabal et al. 2018; Santos et al.
669 2019). In this framework, the contrast between basement inliers strongly reworked in the
670 Neoproterozoic, (i.e. Curitiba Terrane, Camboriú Complex, Encantadas Terrane) and more
671 tectonically stable forelands (LAT and Nico Pérez Terranes) has been expressed in terms of varying

672 metacratonization of the pre-Brasiliano crust (Oriolo et al. 2017; Santos et al. 2019). One
673 paleogeographic reconstruction for the common origin of all these blocks is that they share an
674 affinity with the cratons on the African side of the Pan-African/Brasiliano orogenic systems,
675 particularly the Congo Craton (Oriolo et al. 2016, 2019; Basei et al. 2018; Konopasek et al. 2018;
676 Oyhtançabal et al. 2018). In this model, all of these blocks would share a common geological history
677 in the Paleo- to Mesoproterozoic up to the point, probably in the early Neoproterozoic, in which they
678 were rifted apart. In the model of Basei et al. (2018) this would lead to the generation of oceanic
679 crust and the opening of the Adamastor Ocean. Subsequent closure of this ocean would lead to the
680 collision of the different blocks and a juxtaposition along fault-bound suture zones.

681 Resuming the discussion above on the comparison between the LAT and the Curitiba Terrane and
682 their combined evolution, the recognition of their common ancient history could provide a synthesis
683 in which both models are not necessarily mutually exclusive. Passarelli et al. (2018) highlight that
684 both terranes differ especially in terms of the contrasting ages for the main migmatization event
685 (Paleoproterozoic vs. Neoproterozoic) and structural configuration (NE-SW striking vs. NW-SE
686 striking). These differences can be interpreted as the result of varying degrees of Neoproterozoic
687 overprint even in a collisional orogenic setting, as long as it involves similar fragments that had
688 previously been rifted apart. In this way, both the evidences indicating subduction and preservation
689 of oceanic crust and the similar ancient origin for both terranes could be accommodated.

690

691 **6. Conclusion**

692 The new LA-MC-ICP-MS U-Pb data of zircon and titanite presented in this study offer a complement
693 to the existing geochronological database of the basement of the Luis Alves Terrane. Furthermore,
694 we present the first Hf isotope data of this association, which enables a deeper understanding of its
695 evolution.

696 The critical analysis of the complex U-Pb dataset, supported by Hf analyses, allows a confident
697 interpretation of several magmatic and metamorphic events which formed the LAT. Strongly crustal
698 Hf signatures with ancient model ages are supported by the presence of inherited zircon cores with
699 ages up to 3.2 Ga, giving new constraints on the age of the original extraction of the LAT. Most
700 Archean protoliths were formed at ca. 2.68 Ga, probably during a prolonged magmatic event.
701 Subordinate juvenile accretion took place at ca. 2.5 Ga, supposedly during a second magmatic event,
702 followed by two high-grade metamorphic events at 2.35 and 2.18 Ga responsible for a complete
703 reworking and mixing of the inherited isotopic signatures. A retrograde metamorphic trajectory
704 responsible for the re-equilibration of the metamorphic samples under amphibolite facies conditions
705 is constrained by the crystallization of concordant titanite crystals at 2.02 – 1.99 Ga. Finally, late
706 pegmatitic magmatism is tentatively estimated at between 2.0 and 1.8 Ga.

707 Zircon grains from all four samples show signs of important hydrothermal alteration. Discordant U-Pb
708 spots suggest that the hydrothermal event happened during the Neoproterozoic Brasiliano orogenic
709 cycle. This hydrothermal event did not only produce Pb loss in the dataset but is probably also
710 responsible for the high number of reverse discordant U-Pb ages observed in the dataset by
711 redistributing radiogenic Pb within the crystals.

712 Our data reflects how the LAT remained stable during the Pan-African/Brasiliano orogenic cycle in
713 spite of being surrounded by terranes intensively formed or reworked and/or produced in the Late
714 Neoproterozoic, as a disturbance of the U-Pb system was only achieved through a combination of
715 long-term metamictization and hydrothermalism in the samples. Given the proximity of the studied

716 outcrop with the Neoproterozoic Campo Alegre Basin, it is possible that this hydrothermal overprint
717 was a rather local feature.

718

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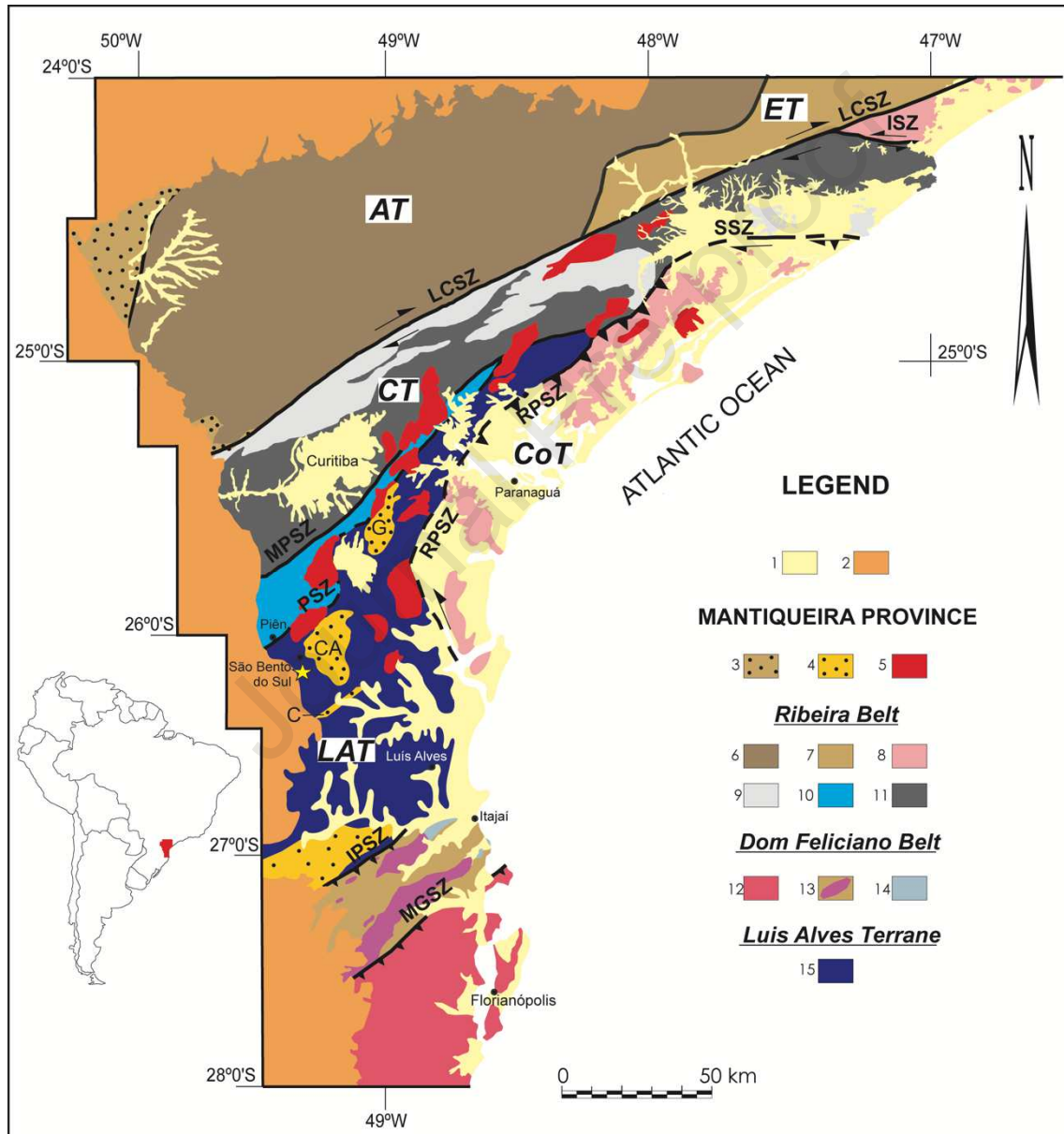
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1043 **Figures and Captions**

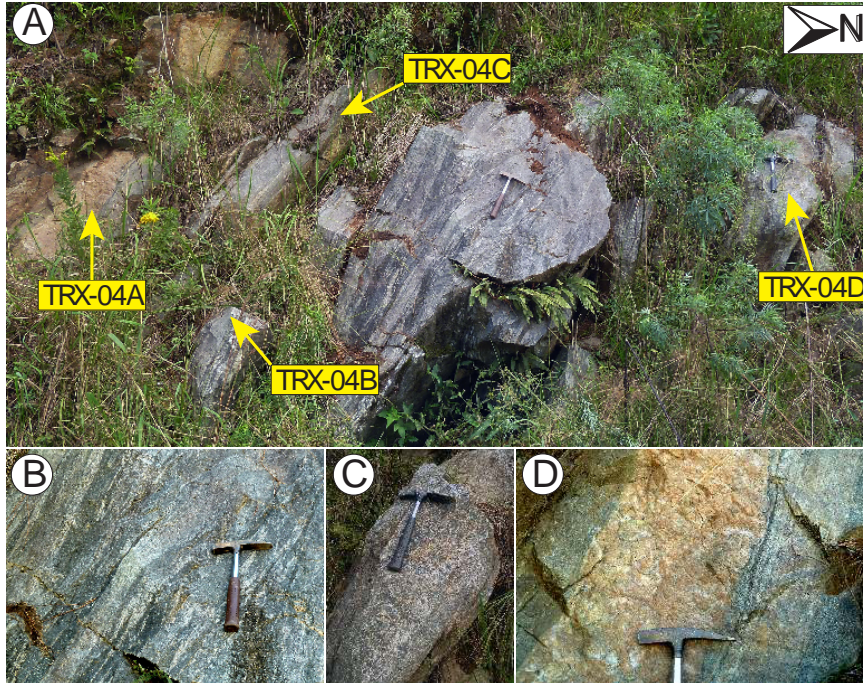


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1045 Fig. 1 –Tectonic and geological sketch of the south-southeast Brazilian Precambrian terranes with sampling
 1046 location of sample TRX-04 (star). 1. Quaternary and Tertiary sediments; 2. Paraná Basin; 3. Eopaleozoic basins;
 1047 4. Neoproterozoic basins (CA: Campo Alegre; C: Corupá; G: Guaratubinha); 5. Graciosa/Serra do Mar Suite
 1048 alkaline granites. **Ribeira Belt**: **Apiaí Terrane (AT)**: 6. Supracrustal and granitic rocks undifferentiated. **Embu
 1049 Terrane (ET)**: 7. Orthogneisses, supracrustal and granitic rocks undifferentiated. **Coastal Terrane (CoT)**: 8.
 1050 Metasedimentary Sequences and gneiss-granitic rocks of Paranaguá/Mongaguá Domains undifferentiated.
 1051 **Curitiba Terrane (CT)**: 9. Supracrustal rocks; 10. Piên Domain: calc-alkaline granitoids and mafic ultramafic
 1052 suite; 11. Atuba-Registro Itatins Complexes. **Dom Feliciano Belt**: 12. Granitoid belt (Florianópolis Batholith); 13.

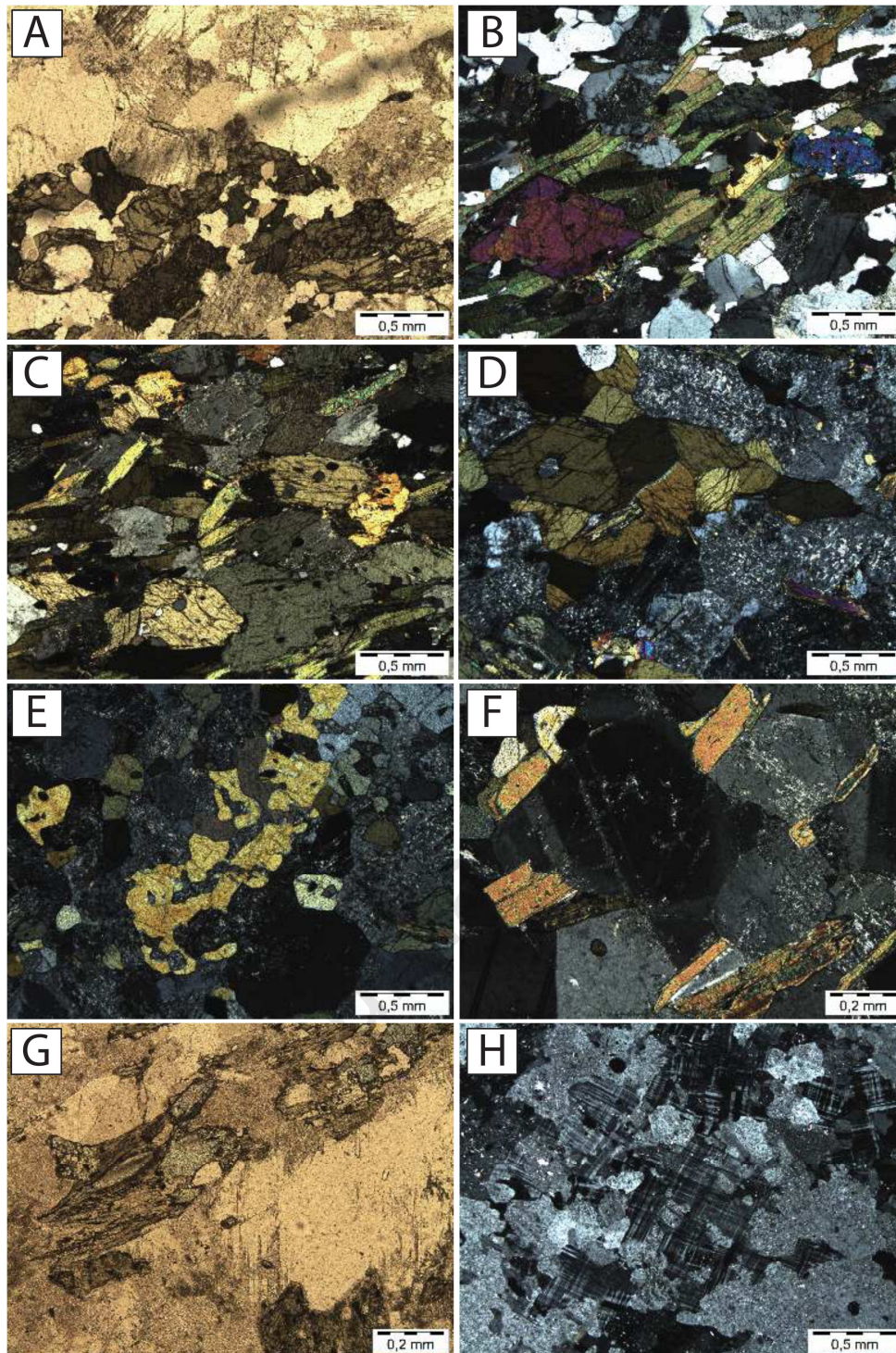
1053 Schist Belt and intrusive granitoids; 14. Basement inliers. ***Luis Alves Terrane (LAT)***: 15. Santa Catarina granulitic
 1054 Complex. **Main shear zones (SZ)**: LCSZ: Lancinha-Cubatão; ISZ: Itariri; SSZ: Serrinha; RPSZ: Rio Palmital; MPSZ:
 1055 Mandirituba-Piraquara; PSZ: Piên; IPSZ: Itajaí-Perimbó; MGSZ: Major Gercino. Modified from Passarelli et al.
 1056 (2018). Based on Basei (2000); Basei et al. (1992), (2000), (2009b); Campos Neto and Figueiredo (1995).

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1059 Fig. 2 – Field pictures of the sampled rocks. (A) Entire outcrop representative of the metamorphic units; the
 1060 yellow arrows indicate the location of the collected samples. (B) orthogneiss in the center of the outcrop.
 1061 Samples TRX-04B and TRX-04B are homogeneous, slightly broader felsic (TRX-04B) and mafic (TRX-04C)
 1062 bands of this gneiss. (C) amphibolite lens, sample TRX-04D. (D) detail picture of the pegmatite, sample TRX-04A.



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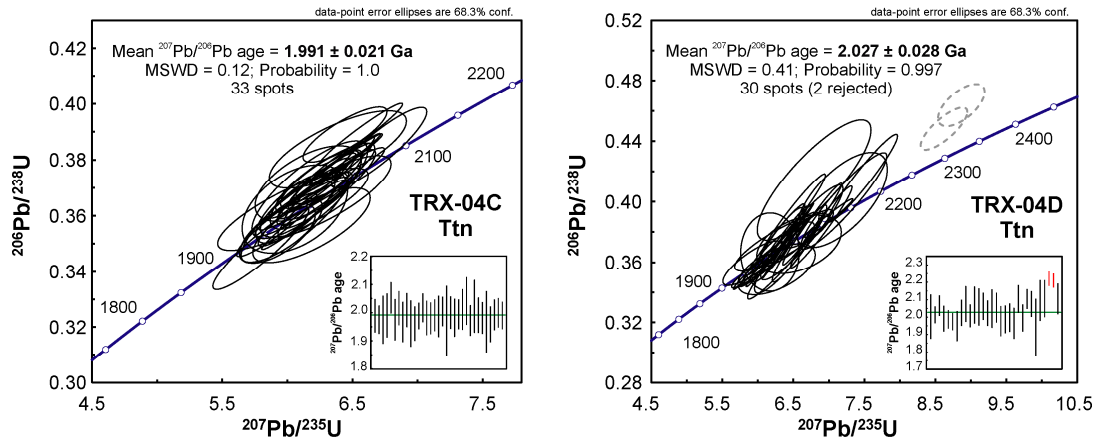
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Fig. 3 – Microphotographs of TRX-04 outcrop samples. **Sample TRX-04B:** (a) Anhedral amphiboles in quartz–feldspatic groundmass of the syenogranite-gneiss TRX-04B. The feldspars look darker because they are very altered. (b): Euhedral epidote with zoning (pink), aligned biotite, feldspar and quartz. **Sample TRX-04C:** (c): Orientated, inclusion rich amphiboles with some biotite and altered feldspar in the quartz-monzonitic gneiss. **Sample TRX-04D (amphibolite):** (d) aggregation of several amphiboles in the middle of very saussuritized plagioclases, (e) Epidote associated to calcite with altered, sometimes tabular plagioclase, (f) The dark plagioclase in the middle shows a hexagonal zoning. Note the 120° angles where 3 grains meet (g) Intergrowth of epidote and biotite. **Sample TRX-04A** (h) Microcline and strongly altered orthoclase (or plagioclase, a clear discrimination is often not possible) in the pegmatite sample TRX-04 A.



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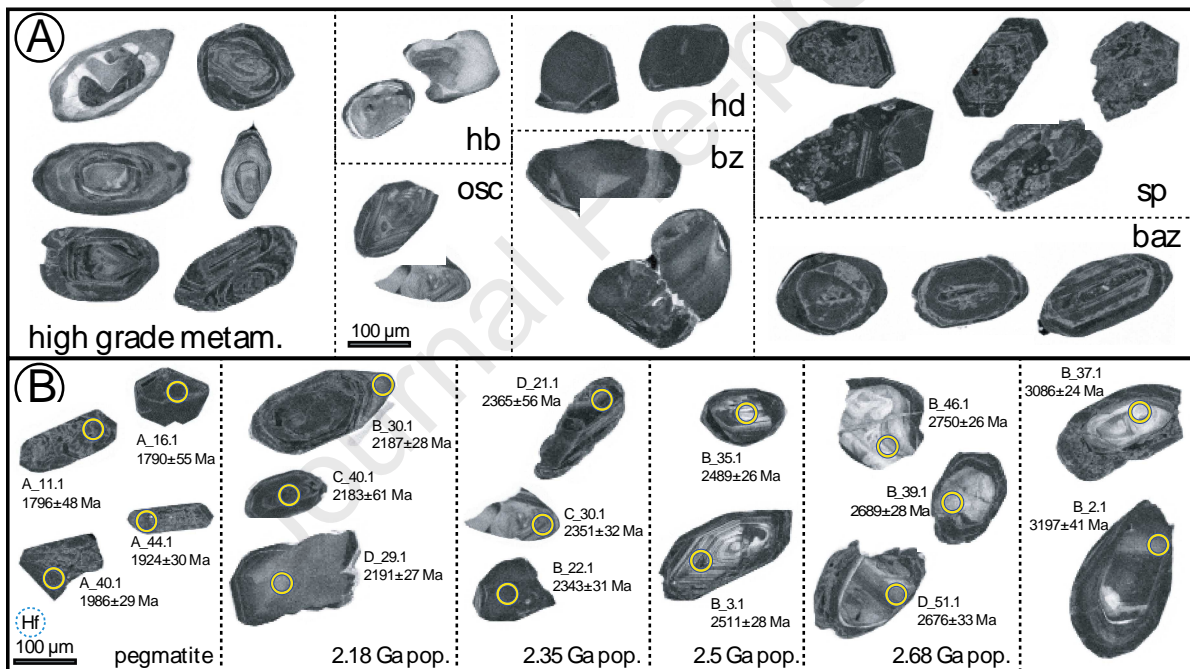
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Figure 4: Concordia diagrams and mean age plots of the titanite U-Pb dataset. Spots used for age calculations are represented as empty black ellipses and empty dashed grey ellipses and red lines in the mean age plot represent discarded analyses.

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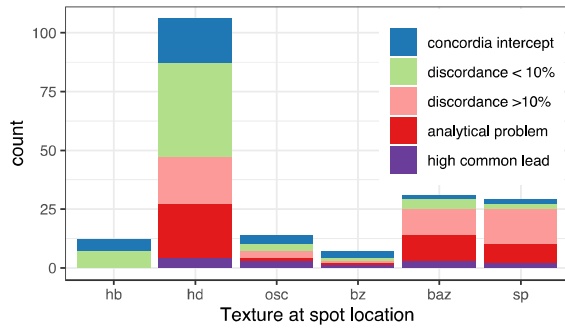
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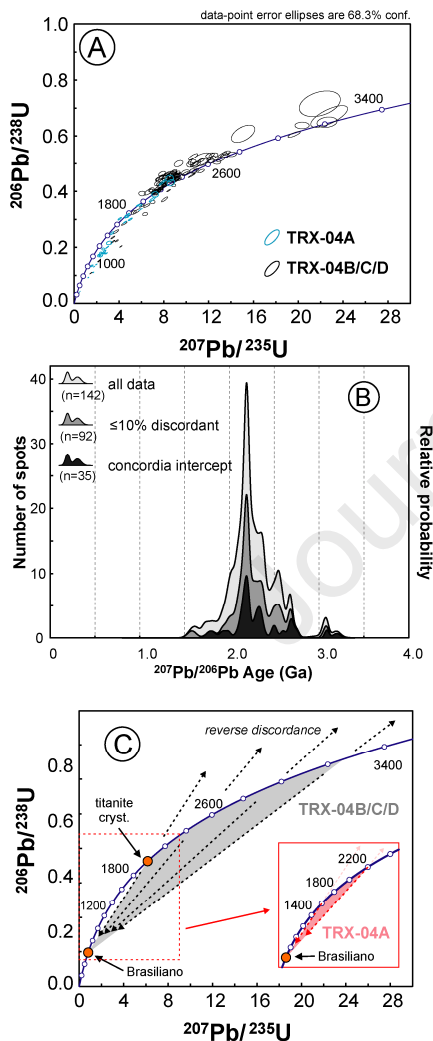
Figure 5: CL images of zircon grains from all samples, scaling is the same in A and B. (A) shows examples of the variety of different textures present the samples. The grains in the left panel show very complex, often chaotic textures typical for high grade metamorphism and cores overprinted by zones of recrystallization or new growth, the others show examples of each classification (hb: homogeneous bright, hd: homogeneous dark, osc: oscillatory zoning, bz: broad zoning, sp: spongy/porous, baz: broad altered zoning). (B) shows highly concordant (intercept with Concordia) grains and ages of each age population. The circles show the 32 μm sized LA spots for the U-Pb analyses. Hf analyses were performed with 47 μm sized LA spots (dashed circle next to scale bar) in the same locations. The corresponding analytical data can be found in the electronic supplementary material.



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1090 Figure 6: Barplot showing the distribution of textures of all samples indicating the proportion of concordant,
 1091 discordant and problematic analyses (baz: broad altered zoning, bz: broad zoning, hb: homogeneous bright, hd:
 1092 homogeneous dark, osc: oscillatory zoning, sp: spongy/porous). Highly discordant (>10%) and problematic
 1093 analyses are mainly related to grains with alteration features such as spongy texture or broad altered zoning.

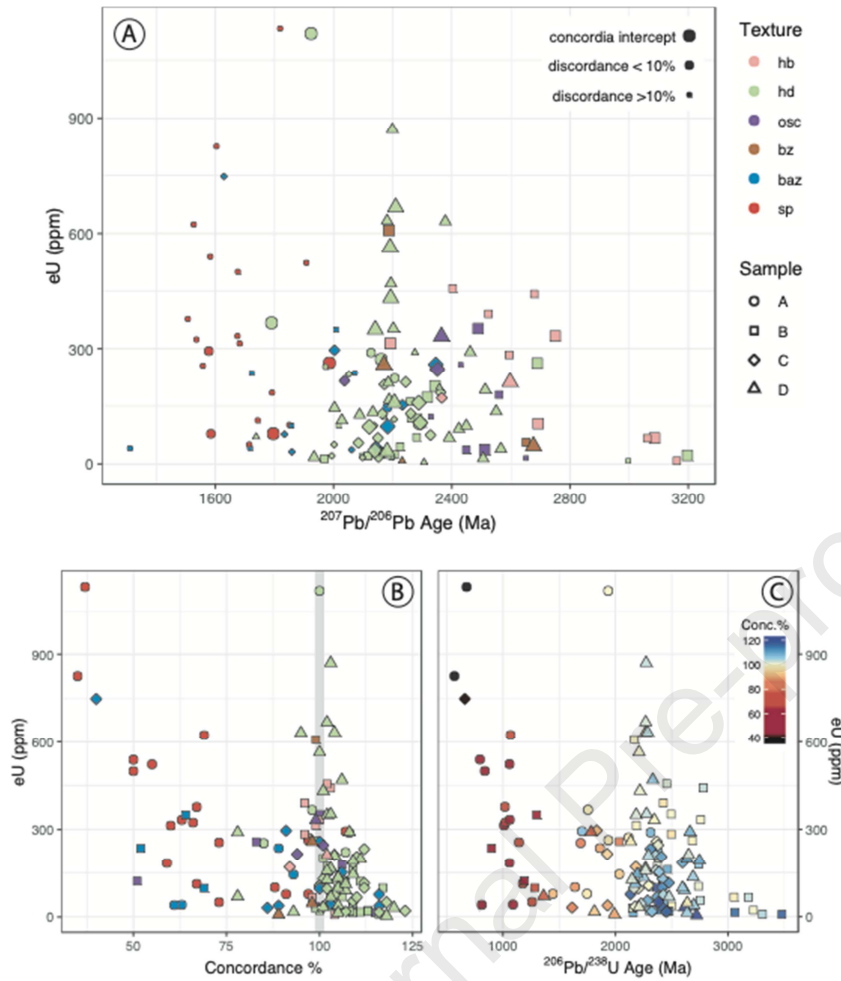
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1096 Figure 7: U-Pb zircon dataset for all spot analyses. The Concordia diagrams (A) shows the dataset comparing
 1097 sample A (blue ellipses) with the other samples (black ellipses). (B) Shows probability density plots according to
 1098 their respective $^{207}\text{Pb}/^{206}\text{Pb}$ ages; the dataset is separated into three categories: spot analyses that intersect the
 1099 Concordia line within uncertainties, analyses $\leq 10\%$ discordant and analyses $> 10\%$ discordant. (C): schematic
 1100 diagram on a Concordia diagram representing how a Neoproterozoic Pb redistribution event on samples
 1101 recording a wide range of geological ages can produce a distribution of results similar to that observed in the
 1102 dataset

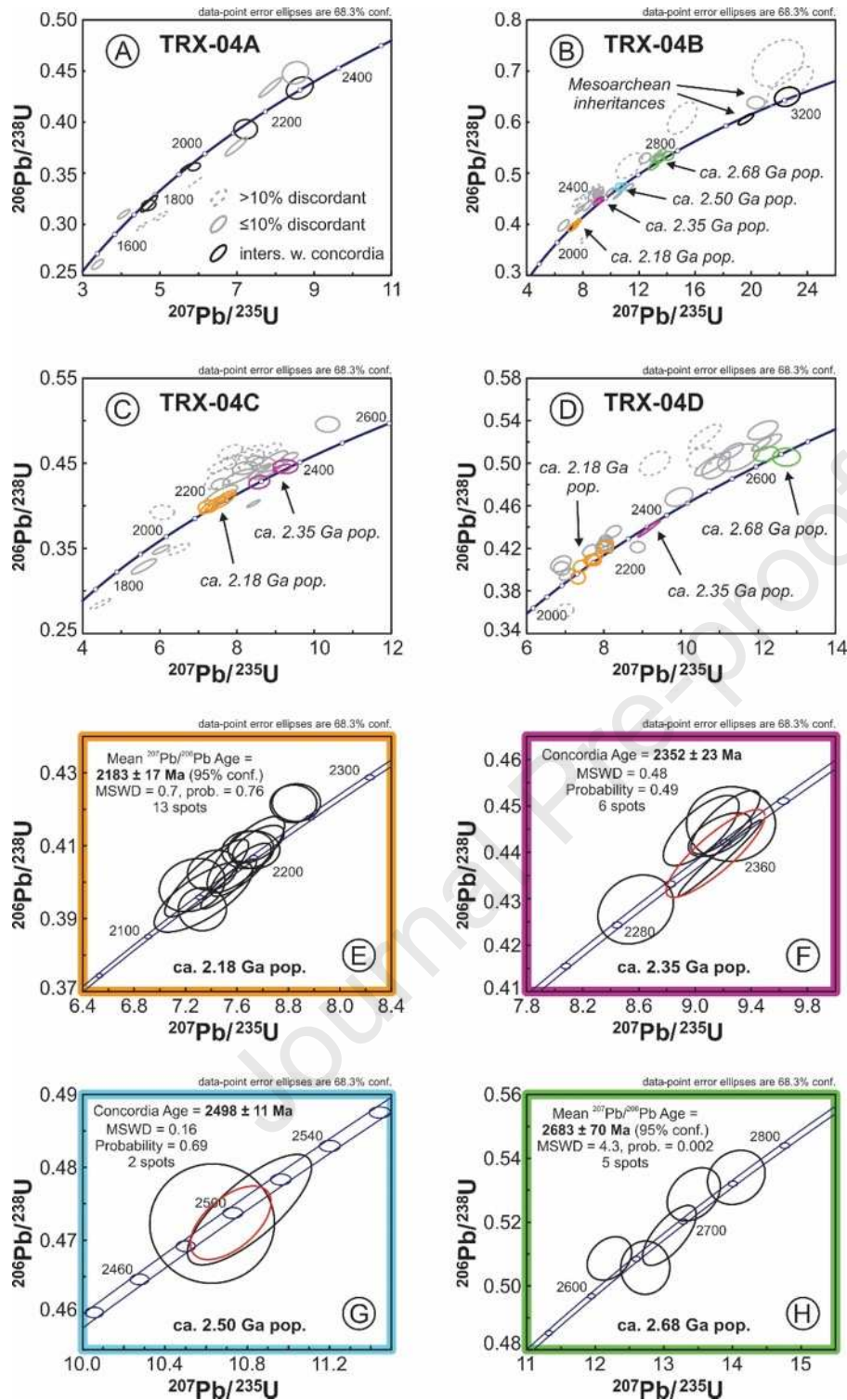
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1105 Figure 8: Relationships of U-Pb ages with effective Uranium ($eU=U + 0.235*Th$). (A) shows the $^{207}\text{Pb}/^{206}\text{Pb}$ Pb ages
 1106 versus eU. Colors indicate the textures (hb: homogeneous bright, hd: homogeneous dark, osc: oscillatory
 1107 zoning, bz: broad zoning, baz: broad altered zoning, sp: spongy/pourous) of the analysed spots, the shapes are
 1108 according to the samples and symbol sizes indicates the degree of concordance. While hb texture is common in
 1109 very old grains, most young and discordant ages have altered textures (sp and baz). (B) shows the eU content
 1110 versus the degree of concordance, colors and shapes are as in A. Most highly normal discordant data (<100%
 1111 Conc) occurs in grains with alteration textures and concordance correlates negatively with eU. Reverse
 1112 discordance (>100%) is common in homogeneous dark grains and equally related with eU. (C) shows $^{206}\text{Pb}/^{238}\text{U}$
 1113 ages versus eU, shapes are as in A, color is according to concordance. For highly normal discordant data eU is
 1114 negatively correlated with the $^{206}\text{Pb}/^{238}\text{U}$ age. For the sake of visibility two grains with $eU > 2000$ ppm, U-Pb ages
 1115 of ~ 1.4 Ga and concordance of $\sim 20\%$ are not displayed in A, B and C.

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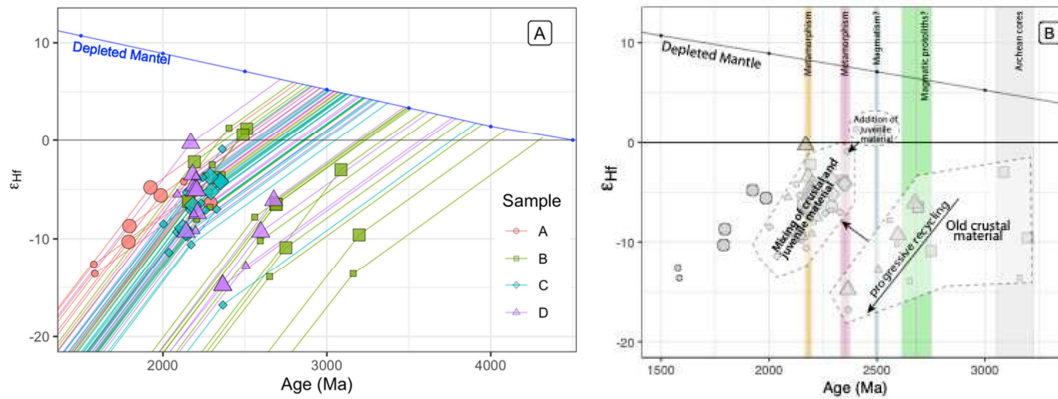
1118 Figure 9: Concordia diagrams highlighting the most concordant U-Pb zircon results for each sample and main
 1119 geological events. A-D: Samples TRX-04A, 04B, 04C and 04D. E-H: Calculated ages for the main populations
 1120 observed in samples TRX-04B, 04C and 04D. In diagrams B-D the crystals used for age calculations are color-
 1121 coded according to the main age populations, and the dataset is represented according to their degree of
 1122 concordance following the legend in diagram A. Diagrams F-G only represent ellipses that intersect the
 1123 Concordia line and calculated Concordia ages (red ellipse).

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1129 Figure 10: Representations of the Hafnium data of samples TRX – 04 A-D. (A): $\epsilon_{\text{Hf}}(t_1)$ values as a function of
 1130 time for the four samples (TRX-04A: red circles, TRX-04B: green squares, TRX-04C: blue diamond, TRX-04D:
 1131 purple triangles). Big symbols correspond to concordant analyses with U-Pb ages intercepting the Concordia
 1132 curve, small symbols correspond to analyses up to 10% discordant. $\epsilon_{\text{Hf}}(t_1)$ is calculated from the measured
 1133 $^{176/177}\text{Hf}$ isotope ratio and the corresponding U-Pb age. The paths indicate the evolution of ϵ_{Hf} as a function of
 1134 time. The intercepts of these paths with the ϵ_{Hf} composition of the depleted mantle (blue line and dots)
 1135 indicate the Depleted Mantle model age T_{DM} . (B) shows the same data as A emphasizing the main zircon
 1136 populations (vertical colored bars, colors and ages are as in Figure 9) and the proposed differentiation and
 1137 mixing processes interpreted for the dataset. Symbols and sizes are as in (A).

1138

- Concordant Zr U-Pb ranging from 3.2 -1.8 Ga represent complex evolution of the LAT
- Titanite crystallized at ≈ 2.0 Ga during a retrograde metamorphic trajectory
- Reverse discordant data indicate hydrothermal alteration during Neoproterozoic
- Hf-isotopes in zircon and titanite U-Pb ages seem to be unaffected by alteration

Journal Pre-proof

Author statement:

Beatrix M.Heller: Investigation, Writing-Original draft, Visualization

Mathias Hueck: Writing-Original draft, Visualization

Claudia R. Passarelli: Writing – Review and Editing

Miguel A.S. Basei: Supervision

Journal Pre-proof

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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