



# Lu-Hf analyses of zircon from the Makoppa Dome and Amalia-Kraaipan area: 1 implications for evolution of the Kimberley and Pietersburg blocks of the Kaapvaal Craton.

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1   **Lu-Hf analyses of zircon from the Makoppa Dome and Amalia-Kraaipan area:**  
2   **implications for evolution of the Kimberley and Pietersburg blocks of the**  
3   **Kaapvaal Craton.**

4

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10    **Abstract**

11    Previously dated zircon crystals from the Amalia-Kraipan granite-greenstone belts  
12    and Makoppa Dome were analysed for their Lu-Hf isotopic characteristics to refine  
13    the geological evolution of these areas. Samples from the Makoppa Dome, belonging  
14    to the Pietersburg Block, largely fall within the epsilon Hf-age range for granitoids  
15    from the eastern part of the block. However, the oldest 3.01-3.03 Ga trondhjemitic  
16    gneisses show that reworking of juvenile mafic crust started earlier in the western  
17    than the eastern part of the block, suggesting a diachronous tectonic evolution. The  
18    three granitoids from the Amalia-Kraipan area fall within the field for Pietersburg and  
19    Kimberley block granitoids. Contribution from older crustal material is seen in a 3.08  
20    Ga schist, likely derived from a volcanic protolith, from the Madibe Belt, in the far east  
21    of the Kimberley Block, with a mantle extraction age of 3.25-3.45 Ga. The data  
22    suggest that the Kimberley Block, like the Pietersburg Block, also contains (minor)  
23    ancient crustal components, derived from a depleted mantle source prior to 3.1 Ga.  
24    The new data suggest that the Kimberley and Pietersburg blocks underwent a very  
25    similar Paleo- to Mesoarchean crustal evolution, with a major crust formation event at  
26    3.1-3.0 Ga followed by successive crust reworking until 2.77 Ga. Lavas of the  
27    Ventersdorp Supergroup, for which zircons from a ca. 2.75 lapilli tuff give  $\epsilon_{\text{Hf}}$  of +2,  
28    are the first evidence of a juvenile source, after 300 Myr of crustal reworking.

29

30    **Introduction**

31    The Kaapvaal Craton is one of the better-studied Archean cratons, thanks to its  
32    relatively accessible and well-exposed geological record. Its division into four  
33    separate terranes or blocks (Eglington and Armstrong, 2004), based on  
34    geochronological data, is now widely accepted (Figure 1), with the Swaziland Block  
35    having attracted the most academic attention as it hosts the craton's oldest rocks at  
36    3.66-3.70 Ga (Compston and Kröner, 1988; Kröner et al., 1996; Robb et al., 2006;  
37    Zeh et al., 2011). Recorded ages are somewhat younger in the Witwatersrand and

38 Pietersburg blocks (Anhaeusser, 2019) at  $\leq$ 3.34 Ga (Laurent and Zeh, 2015; Poujol  
39 and Anhaeusser, 2001). Because of its extensive cover, the Kimberley Block is the  
40 least known, and virtually no ages older than ca. 3.2 Ga have been found  
41 (Anhaeusser and Walraven, 1999; Cornell et al., 2018; Poujol et al., 2008). The  
42 boundaries between the four terranes are based on geophysically defined  
43 lineaments, which may or may not have any surface expression, as well as  
44 recognisable faults and shear zones, and extensions thereof (Eglington and  
45 Armstrong, 2004). As such, they are somewhat dependent on changing  
46 interpretations of the geophysical data (Corner and Durrheim, 2018), additional  
47 geochronological information and also geochemical data. In the latter respect, the  
48 development of the past fifteen years of laser ablation multi-collector inductively  
49 coupled mass spectrometry (LA-MC-ICPMS), which permits the determination of the  
50 Hf isotopic characteristics of the same zircon grains that provide the age of  
51 intrusions, needs to be mentioned. Zircon is the most robust carrier of U-Pb age data,  
52 and has therefore been the mineral of choice to obtain reliable geochronological  
53 information. Zircon's high contents of the element hafnium, a geochemical twin of  
54 zirconium, and limited amount of the radioactive parent element lutetium, makes  
55 them also the best carrier of Hf isotopic information. As mantle and crust have  
56 contrasting Lu/Hf ratios, they develop different Hf isotope ratios over time. Studying  
57 the Hf isotopic compositions of dated zircon crystals therefore allows us to test  
58 whether intrusions with the same age also have similar magma sources; and whether  
59 these sources were juvenile (mantle-derived), or incorporated older crustal materials.  
60 As isotopic information, unlike whole rock geochemistry, is only affected by the  
61 sources contributing to the magma, irrespective of crystal fractionation, it is a robust  
62 test for similarity among rocks of the same age. This technique has been applied  
63 successfully to a number of plutons of the Kaapvaal Craton (see overviews in Kröner  
64 et al., 2019 and Laurent et al., 2019, and the many works by Zeh and coworkers (e.g.  
65 2009, 2011)), but a great number of intrusives of which the age is known has not

66 been analysed for the Hf isotopic composition. In addition to providing information on  
67 the petrogenesis of the igneous rocks from which the zircon grains are derived,  
68 establishing an age-Hf isotopic zircon database for the Kaapvaal Craton is also  
69 important for the study of detrital zircon in sedimentary rocks, which provides  
70 information on the sediments' provenance.

71 To add to the available information on the geological evolution of the Kimberley and  
72 Pietersburg blocks and Kaapvaal Craton zircon age-Hf isotopic database, we  
73 performed Lu-Hf isotopic analyses on zircon samples of ten igneous rocks from the  
74 Amalia-Kraipan area of the Kimberley Block and the Makoppa Dome of the  
75 Pietersburg Block that were analysed for their U-Pb characteristics previously  
76 (Anhaeusser and Poujol, 2004; Poujol et al., 2002; Poujol et al., 2008; Poujol et al.,  
77 2005). Our new data are compatible with the interpretation that the Makoppa Dome is  
78 indeed part of the Pietersburg Block, and provides the earliest evidence for reworking  
79 of juvenile mafic material in the region, whereas they add to our relatively poor  
80 knowledge of the Kimberley Craton. Additionally, these data are compared to the  
81 existing detrital zircon database for Archean-Paleoproterozoic Kaapvaal Craton and  
82 provide matches for zircon grains from the Witwatersrand Supergroup and Waterberg  
83 Group.

84

## 85 **Geological background**

86 The geology of the Pietersburg Block has mainly been studied in its eastern part,  
87 between the extension of the Palala Shear Zone and the Thabazimbi-Murchison  
88 Lineament (Figure 1c), which are interpreted to be the northern and southern  
89 boundary, respectively, of the Pietersburg Block (Eglinton and Armstrong, 2004).  
90 The Southern Marginal Zone of the Limpopo Belt, which is located north of the Hout  
91 River Shear Zone (Figure 1c), is also deemed part of the Pietersburg Block  
92 (Eglinton and Armstrong, 2004). The western part of the block, in which the  
93 Makoppa Dome is located, is separated from the eastern part by the ca. 2.05 Ga

94 Bushveld Complex, which straddles the boundary of the Witwatersrand and  
95 Pietersburg blocks. In its eastern part, the Pietersburg Block consists of ENE-WSW  
96 running greenstone belts (the Murchison, Giyani and Pietersburg belt), with several  
97 generations of granitoids. The oldest of these are the Goudplaats-Hout River tonalite-  
98 trondhjemite-granodiorite (TTG) gneisses at 3.2-3.43 Ga (Laurent and Zeh, 2015).  
99 Further TTG intrusions followed from 3.0 to ca. 2.8 Ga, and more K-rich magmatism  
100 from 2.9 to 2.67 Ga (see Laurent et al., 2019 for a review). Magmatism has been  
101 interpreted to reflect accretion of several arcs, and a >3.2 Ga continental nucleus, to  
102 the proto-Kaapvaal Craton, consisting of the Swaziland and Witwatersrand blocks.  
103 The Makoppa Dome straddles the boundary between South Africa and Botswana,  
104 with an area of ca. 7600 km<sup>2</sup>. The only published work on the area is the paper by  
105 Anhaeusser and Poujol (2004), which also provides the ages of the zircon used for  
106 the present study. The rocks of the Makoppa Dome are largely obscured by post-  
107 Mesozoic sediments, but consist of both granitoids and greenstones (Figure 1b). The  
108 latter are amphibolites, serpentinite, talc/chlorite schists and banded iron formations.  
109 The granitoids, which were the focus of the U-Pb zircon and whole rock geochemistry  
110 study by Anhaeusser and Poujol (2004), are the Vaalpenskraal trondhjemite/tonalite  
111 gneiss, the Makoppa granodiorite/monzogranite and the Rooibokvlei  
112 granodiorite/monzogranite. Intrusion and deformation relationships define the  
113 Vaalpenskraal gneisses to be the oldest felsic rocks in the area.  
114 The Kimberley Block of the Kaapvaal Craton has outcrops of Archean rocks only in  
115 the Kraipan-Amalia area, and the area around Marydale. Granitoids from the latter  
116 area were recently subject of study by Cornell et al. (2018), who reported ages of ca.  
117 2.95 to 2.72 Ga, and xenocrysts up to 3.18 Ga. This work also quotes unpublished  
118 ages up to 3.28 Ga from basement samples retrieved during diamond mining. This is  
119 similar to the ca. 3.25 Ga unpublished ages quoted by Drennan et al. (1990) for  
120 areas west of the Colesberg Lineament, which forms the boundary with the  
121 Witwatersrand Block. The Kraipan area contains greenstone outcrops that occur as

122 three discontinuous NNW-trending belts, the Stella, Kraaipan and Madibe belts, from  
123 W to E (Brandl et al., 2006). The greenstones of the Amalia area lie along the  
124 extension of the Stella Belt to the south (Figure 1a), and contain interbedded  
125 pyroclastic rocks; the discontinuous outcrops may represent tectonically juxtaposed  
126 slivers (Brandl et al., 2006). Granitoids, some containing greenstone remnants, occur  
127 in the Amalia and Kraaipan areas, but intrusive relationships are unclear because of  
128 poor exposures. The rocks of the Amalia area are overlain by Ventersdorp  
129 Supergroup Lavas, dated at  $2729 \pm 3$  Ma (Poujol et al., 2005).

130

### 131 **Sample description and ages**

132 Table 1 gives an overview of the samples analysed. All background information,  
133 including cathodoluminescence images, can be found in the referenced papers.

### 134 **Pietersburg Block**

135 The five samples from the Makoppa Dome, in the western part of the Pietersburg  
136 Block, have been described by Anhaeusser and Poujol (2004), and the following is  
137 based on their findings. All samples have undergone some degree of sericitisation,  
138 but no other evidence of metamorphism was noted, apart from the gneissosity of the  
139 oldest samples.

140 Samples MK10 and MK11 were taken from the Vaalpenskraal trondhjemite gneiss,  
141 along the Crocodile River in the north central part of the dome (Figure 1b). The zircon  
142 crystals are yellowish to pink in colour, elongated to prismatic, with some rounded  
143 edges. Cathodoluminescence imaging showed apparent core-rim structures, with  
144 brighter cores and darker rims; rare crystals showed simple igneous zoning. Dating  
145 of sample MK10 was done by LA-ICP-MS and yielded a weighted average  
146  $^{207}\text{Pb}/^{206}\text{Pb}$  age of  $3013 \pm 11$  Ma, measured mostly on the bright cores. This was  
147 interpreted to represent the age of intrusion. Zircon rims gave a poorly constrained  
148 upper intercept age of ca. 2.6 Ga, interpreted to reflect a later metamorphic event.

149 Sample MK11 was also dated by LA-ICP-MS, but gave poor results with an older  
150 age, likely igneous, of  $3034 \pm 64$  Ma, and a likely resetting event at  $2842 \pm 35$  Ma.  
151 Sample MK5 represents the Makoppa granodiorites/monzogranites and was taken  
152 near the town of the same name. These relatively large, yellow to amber crystals,  
153 showed simple zoning in CL imaging. Zircon crystals from this sample were analysed  
154 by ID-TIMS and define an upper intercept age of  $2886 +3/-2$  Ma, which was  
155 interpreted to reflect the intrusive age of this phase.  
156 Samples MK2 and MK3 represent the Rooibokvlei granodiorite/monzogranites. MK2  
157 yielded pink to metamict zircon grains, of which many showed complex zoning in CL  
158 imaging, but few crystals with simple igneous zoning were also present. ID-TIMS  
159 dating yielded an upper intercept age of  $2777 \pm 2$  Ma. This was confirmed by LA-ICP-  
160 MS dating, which yielded an upper intercept age of  $2777 \pm 35$  Ma. Many of the more  
161 complexly zoned crystals showed much younger ages, trending towards a lower  
162 intercept of ca. 800 Ma. Sample MK3 yielded crystals that were similar in character to  
163 those of MK2, and ID-TIMS dating gave an upper intercept age of  $2797 \pm 2$  Ma. This  
164 is regarded as the emplacement age of this granitoid.

165

### 166 ***Kimberley Block***

167 All five samples analysed come from the Kraaipan-Amalia area of the Kimberley  
168 Block. Three of them (Madibe1, MAD1 and KP5; Poujol et al., 2002; Poujol et al.,  
169 2008) come from the Kraaipan area (Figure 1c); these samples show limited  
170 sericitisation and epidotisation (Anhaeusser and Walraven, 1999). Two others (AL3,  
171 BOT1; Poujol et al., 2002; Poujol et al., 2005) come from near Amalia.  
172 Sample Madibe-1 (Poujol et al., 2008) is a lower greenschist-facies quartz-sericite  
173 schist that has been interpreted, based on its whole rock geochemistry, as a calc-  
174 alkaline lava from the Madibe belt in the easternmost part of the Kraaipan area. The  
175 sample was taken in the hanging wall of the orebody of the Madibe gold mine. This  
176 borehole sample is located very close to the Colesberg Lineament (Figure 1c), which

177 is interpreted to run through Mahikeng (McCarthy et al., 2018). Zircon crystals in this  
178 drill core sample were rare, but those present are euhedral and have well-defined  
179 oscillatory zoning. Eight concordant SIMS analyses yielded a weighted average  
180  $^{207}\text{Pb}/^{206}\text{Pb}$  emplacement age of  $3083 \pm 6$  Ma, whereas one older xenocrystic grain  
181 gave a concordant  $^{207}\text{Pb}/^{206}\text{Pb}$  date of  $3201 \pm 4$  Ma.

182 Sample KP5 (Poujol et al., 2002), a pink granodiorite, was taken near the Kraipan  
183 railway siding. Zircon crystals are prismatic, and show well-developed zoning in  
184 cathodoluminescence (CL) imaging. The ion microprobe analyses gave a weighted  
185 mean  $^{207}\text{Pb}/^{206}\text{Pb}$  age of  $2913 \pm 17$  Ma for the emplacement of this granodiorite,  
186 without any evidence of inheritance.

187 Sample MAD1 was taken near the town of Madibogo and consists of a grey  
188 granodiorite. Zircon grains look similar to those in KP5, and their ion microprobe  
189 emplacement age is also indistinguishable at  $2917 \pm 9$  Ma. Some grains appear to  
190 have suffered Pb-loss with  $^{207}\text{Pb}/^{206}\text{Pb}$  apparent ages down to  $2629 \pm 23$  Ma.

191 Sample BOT1 was taken between Amalia and Schweizer-Reneke (Poujol et al.,  
192 2005) and consists of a little-deformed accretionary lapilli tuff, which still contains  
193 fresh volcanic glass (Jones and Anhaeusser, 1993). Two types of zircon crystals  
194 were extracted: prismatic, euhedral zircon with simple, CL-dark internal structures,  
195 and smaller, more rounded grains with a very bright CL-response. When analysed by  
196 ion microprobe, the latter type gave Mesoproterozoic dates ( $1038 \pm 48$  Ma and  $1538$   
197  $\pm 50$ ), and the former a weighted mean  $^{207}\text{Pb}/^{206}\text{Pb}$  age of  $2754 \pm 5$  Ma interpreted as  
198 the emplacement age.

199 Sample AL3 (Poujol et al., 2002) was taken to the SSE of Amalia and represents a  
200 fine-grained leuco-trondjemite gneiss, interpreted to be representative of the earliest  
201 basement in this area. Zircon crystals are euhedral to subhedral, pink, and CL  
202 imaging shows zoning and potential core-rim structure. Ion microprobe analyses,  
203 however, show a largely homogeneous age group with a weighted mean  $^{207}\text{Pb}/^{206}\text{Pb}$   
204 emplacement age of  $2939 \pm 10$  Ma, although Pb-loss towards a lower intercept

205 around 1.25 Ga was also observed. Only one core gave a near-concordant  
206  $^{207}\text{Pb}/^{206}\text{Pb}$  apparent age of  $3178 \pm 10$  Ma.

207

208 More detailed descriptions of the samples, as well as CL images of the zircon  
209 crystals, are given in the cited publications.

210

## 211 **Analytical techniques**

212 LA-MC-ICPMS analyses for Lu-Hf were done on the same mounts that were imaged  
213 in the course of the geochronological investigations. The images and age results  
214 were used as a guide for the ablations, but not all zircon grains used for the study  
215 had been analysed for their U-Pb isotopic characteristics. The  $^{207}\text{Pb}/^{206}\text{Pb}$  ages  
216 quoted in the previous section were used for the calculation of initial isotope ratios.

217 The Lu-Hf analyses were done at the Department of Geology, University of  
218 Johannesburg, using a NPII MC-ICP-MS and ASI Resolution laser ablation system.

219 The spot size used was 50-70  $\mu\text{m}$ , and ablations were done at a repetition rate of 6-7  
220 Hz; these parameters were adjusted to match the size and expected depth of the  
221 domain to be ablated. The fluence was ca. 4.6 J/cm<sup>2</sup>. For details on the data  
222 reduction, including interference corrections, see Jacobs et al. (2017). Reference  
223 materials analysed during the course of the study gave results within uncertainty  
224 (quoted at the 2 sigma level) of published values for  $^{176}\text{Hf}/^{177}\text{Hf}$  (Mud Tank: 0.282495  
225  $\pm 0.000018$ , n=16; TEM-2:  $0.282658 \pm 0.000034$ , n=13; LV11:  $282814 \pm 0.000016$ , n=  
226 8). Additionally, the quality of the analyses for the unknowns was monitored by  
227 measuring their invariant  $^{178}\text{Hf}/^{177}\text{Hf}$  and  $^{174}\text{Hf}/^{177}\text{Hf}$  ratios, which yielded averages of  
228  $1.467277 \pm 0.000053$  and  $0.008661 \pm 0.000060$ , respectively. All results can be  
229 found in Electronic Supplement 1.

230 The measured  $^{176}\text{Hf}/^{177}\text{Hf}$  ratios for the unknowns were calculated to their initial value  
231 using the measured  $^{176}\text{Lu}/^{177}\text{Hf}$  ratios, a  $^{176}\text{Lu}$  decay constant of  $1.867 \times 10^{-11}$  year<sup>-1</sup>  
232 (Scherer et al., 2007) and the emplacement ages obtained during previous studies

233 (Table 1). The calculations of epsilon Hf were done with a present-day chondritic  
234  $^{176}\text{Hf}/^{177}\text{Hf}$  value of 0.282785 and  $^{176}\text{Lu}/^{177}\text{Hf}$  of 0.0336 (Bouvier et al., 2008).  
235 Depleted mantle parameters are  $^{176}\text{Hf}/^{177}\text{Hf}$  0.28325 and  $^{176}\text{Lu}/^{177}\text{Hf}$  of 0.0388,  
236 following Griffin et al. (2000).

237

## 238 **Results**

239 Full results can be found in Electronic Supplement 1; averages are given in Table 1.

### 240 **Pietersburg Block**

241 For trondhjemite gneiss sample MK10, twelve ablations were performed, with two  
242 targeting younger rims. The ten analyses on the igneous cores gave  $^{176}\text{Hf}/^{177}\text{Hf}_{3013}$   
243 0.28094-0.28099 (Figure 2a), corresponding to a  $\varepsilon\text{Hf}_{3013}$  of +3.5 to +5.5 (Figure 2b),  
244 with an average (mean) of  $+4.1 \pm 1.2$  (2 standard deviations). The two younger rims  
245 gave only marginally higher initial Hf isotope ratios.

246 Ten zircon grains were analysed for the other sample of trondhjemite gneiss, MK11,  
247 from the same area, and the results are consistent with the other sample, giving an  
248 average  $\varepsilon\text{Hf}_{3034}$  of  $4.9 \pm 1.3$ .

249 Sample MK5 from the Makoppa monzogranite is represented by ten analyses.  
250  $^{176}\text{Hf}/^{177}\text{Hf}_{2886}$  varies from 0.28093 to 0.28101, corresponding to  $\varepsilon\text{Hf}_{2886}$  of 0 to +3.0,  
251 with an average of  $+1.6 \pm 1.9$ .

252 Twelve zircon crystals were analysed for the MK3 Rooibokvlei monzogranite. These  
253 give  $^{176}\text{Hf}/^{177}\text{Hf}_{2797}$  0.28095-0.28105, or  $\varepsilon\text{Hf}_{2797}$  -1.2 to +2.2, and an average  $\varepsilon\text{Hf}_{2797}$   
254  $+0.1 \pm 2$ .

255 Ten analyses were done on the other Rooibokvlei monzogranite sample, MK2, of  
256 which the eight analyses with igneous ages gave results indistinguishable from  
257 sample MK3, with average  $\varepsilon\text{Hf}_{2777}$  -0.4  $\pm 1.5$ . The two areas with younger  $^{207}\text{Pb}/^{206}\text{Pb}$   
258 apparent ages are indistinguishable in their initial  $^{176}\text{Hf}/^{177}\text{Hf}$  ratios from the older  
259 grains.

260

261 ***Kimberley Block***

262 For quartz-sericite schist sample Madibe-1, fourteen zircon crystals were analysed,  
263 with one spot per zircon. This also included the one older zircon xenocryst at 3.2 Ga.  
264 Excluding the latter, 12 grains gave initial  $^{176}\text{Hf}/^{177}\text{Hf}$  values of 0.28077 to 0.28089,  
265 which translates into epsilon- $\text{Hf}_{3083}$  values of -0.8 to +3 (Figure 2a,b). The one dated  
266 older zircon gave a lower  $^{176}\text{Hf}/^{177}\text{Hf}_{3201}$  of 0.280728, corresponding to  $\varepsilon\text{Hf}_{3201} = +0.4$ .  
267 One of the undated grains gave a similarly low value, and could therefore also  
268 represent an older grain. The average of the 12 zircon grains is  $\varepsilon\text{Hf}_{3083} +1.2 \pm 3$   
269 (2SD).

270 For granodiorite sample KP5, eleven analyses were performed, giving  $^{176}\text{Hf}/^{177}\text{Hf}_{2913}$   
271 from 0.28096 to 0.28100, and a range of  $\varepsilon\text{Hf}_{2913}$  of +1.8 - +4.5. The average is  $\varepsilon\text{Hf}_{2913}$   
272 of  $+2.8 \pm 1.5$ .

273 Fourteen analyses were done on zircon from granodiorite sample MAD1, including  
274 two analyses on a single grain, of which the rim had given a younger age. Excluding  
275 this grain, twelve analyses gave  $^{176}\text{Hf}/^{177}\text{Hf}_{2917}$  from 0.28092 to 0.28099, or  $\varepsilon\text{Hf}_{2917}$  of  
276 +0.6 - +3.0, resulting in an average  $\varepsilon\text{Hf}_{2917}$  of  $+2.8 \pm 1.5$ , which is indistinguishable  
277 from the previous sample. For the grain that had given a ca. 2.63 Ga rim age, both  
278 core and rim gave somewhat lower initial Hf isotopic values, indicating that there  
279 might have been some overlap between the two domains during the analyses.

280 For accretionary lapilli tuff sample BOT1 from the Amalia area, thirteen zircon  
281 crystals were analysed. Ten of these constitute a homogeneous group, with  
282  $^{176}\text{Hf}/^{177}\text{Hf}_{2754}$  0.28106-0.28110, or  $\varepsilon\text{Hf}_{2754}$  of +1.6 - +3.2, with average  $\varepsilon\text{Hf}_{2754}$  of +2.3  
283  $\pm 1.0$ . Three undated grains gave much higher  $^{176}\text{Hf}/^{177}\text{Hf}$  ratios (0.2817-0.2824) and  
284 are assumed to be part of the Mesoproterozoic population (ca. 1-1.5 Ga) described  
285 by Poujol et al. (2005). As these analyses cannot give any information on the

286 Archean evolution of the Kaapvaal Craton, which is the subject of this paper, they will  
287 not be discussed further.

288 Sixteen grains were analysed from leuco-trondhjemite sample AL3. Thirteen of these  
289 cluster at  $^{176}\text{Hf}/^{177}\text{Hf}_{2939}$  0.28096-0.28101, corresponding to  $\varepsilon\text{Hf}_{2939}$  of +2.4 - +4.4,  
290 with an average of  $+3.2 \pm 1$ . The one older xenocryst gave significantly lower  
291  $^{176}\text{Hf}/^{177}\text{Hf}_{3178}$  of 0.28087 ( $\varepsilon\text{Hf}_{3178} = +5$ ). Two undated grains also give substantially  
292 lower  $^{176}\text{Hf}/^{177}\text{Hf}$  than the main group (0.28084), so zircon grains with older ages  
293 could potentially be more common in sample AL3 than detected by Poujol et al.  
294 (2002).

295

296

297 **Discussion**

298 To facilitate the interpretation of the data, the average epsilon Hf values for the ten  
299 analysed samples are shown in Figure 3a, together with similarly averaged literature  
300 data. The two older xenocystic cores are shown separately. In this diagram, all other  
301 Kimberley Block Hf isotopic data points are from Cornell et al. (2018), whereas the  
302 Pietersburg Block data are those used in the recent review paper by Laurent et al.  
303 (2019). All references to the literature data are given in Electronic Supplement 2.

304

305 **Pietersburg Block**

306 The general geochemistry and ages of the granitoids of the Makoppa Dome in the  
307 Pietersburg Block were found to be similar to the data from the area to the east of the  
308 Bushveld Complex (Laurent et al., 2019). Our new zircon Hf isotope data confirm this  
309 finding for the ca. 2.9-2.8 Ga samples from the Rooibokvlei (MK2, 3) and Makoppa  
310 (MK5) granodiorite/monzogranite. These three samples overlap in terms of age and  
311 Hf isotopic composition with the various biotite-(muscovite) granites from the eastern  
312 part of the Pietersburg Block. The two samples from the Vaalpenskraal trondhjemitic

313 gneisses are, however, slightly different. Apart from three samples of tonalite-  
314 trondhjemite-granodiorite (TTG) of 3.35-3.25 Ga that have been interpreted to form  
315 the nucleus of a separate ‘crustal nucleus’ (Laurent et al., 2019), the Vaalpenskraal  
316 trondhjemite is the oldest TTG in the Pietersburg Block of which Hf isotope data have  
317 been obtained. The samples are slightly older than the ca. 2.97 Free State Tonalite  
318 of the Rooiwater Complex and Rubbervale felsic volcanics, both in the Murchison  
319 Belt, for which the strongly superchondritic data were reported by Zeh et al. (2013).  
320 Similarly, the Vaalpenskraal trondhjemitic gneisses also record initial epsilon Hf<sub>3.0Ga</sub>  
321 values of +4 - +5. These age-epsilon Hf values overlap with a single older zircon  
322 reported in a 2.88 Ga trondhjemite sampled just north of the Rooiwater Complex by  
323 Laurent and Zeh (2015). A slightly older zircon xenocryst (at 3.115 Ga) with an even  
324 higher initial epsilon Hf value (+6) was reported by the same authors from a 2.83 Ga  
325 biotite granite north of the Hout River Shear zone.  
  
326 The trend in the linear age-epsilon Hf array for the eastern part of the Pietersburg  
327 Block has been interpreted by Laurent and Zeh (2015) and Laurent et al. (2019) as  
328 reflecting partial melting of depleted mantle at 3.15-2.97 Ga, followed by reworking of  
329 the mafic rocks to TTGs at 2.97-2.88 Ga. Subsequently, the TTG-type crust with a  
330  $^{176}\text{Lu}/^{177}\text{Hf}$  ratio of 0.0022 was internally further transformed into grey gneisses and  
331 biotite-granites, with only very minor contributions from older crust. Our new data  
332 from the Makoppa Dome show that formation of felsic crust (Vaalpenskraal  
333 trondhjemite, with SiO<sub>2</sub> contents of 69-75%, Anhaeusser and Poujol, 2004) already  
334 started 3.03-3.01 Ga ago. Assuming a mafic protolith with  $^{176}\text{Lu}/^{177}\text{Hf}=0.022$ , the Hf  
335 isotope data give a mantle extraction age of 3.15-3.10 Ga (Figure 3a, Table 1), in  
336 agreement with the estimate by Laurent et al. (2019). The three samples from the  
337 two younger granitoids fall within the  $^{176}\text{Lu}/^{177}\text{Hf}=0.0022$  array that envelops the  
338 Pietersburg Block intrusives from the eastern side. All five samples fall towards the  
339 lower part of the array, in agreement with the old mantle extraction age of the earliest  
340 samples compared to those from the eastern side of the block. Note that none of the

341 samples fall below the array, which would reflect a contribution of older continental  
342 crust (Figure 3a). This is in agreement with their distal position relative to the old  
343 continental nucleus on the eastern side, and the lack of older inherited zircon.

344 The average  $^{176}\text{Lu}/^{177}\text{Hf}$  ratio of the zircon crystals is plotted in Figure 3b (in contrast  
345 to the previously discussed  $^{176}\text{Lu}/^{177}\text{Hf}$  ratios of the protoliths that define the arrays),  
346 showing that our samples also plot in the same field as previously analysed zircon  
347 grains from the Pietersburg Block. The two younger samples, of the Rooibokvlei  
348 monzogranite have higher  $^{176}\text{Lu}/^{177}\text{Hf}$  than the other three samples, which matches  
349 their whole rock composition, with Lu concentrations 3-8 times higher in the  
350 Rooibokvlei samples (Anhaeusser and Poujol, 2004). This adds to recent discussions  
351 whether zircon trace element data can be used to determine the composition of the  
352 magma it crystallised from (e.g. Chapman et al., 2016; Reimink et al., 2020). In this  
353 case, this appears to hold, on the assumption that the whole rock compositions are  
354 representative for the liquid composition.

355 Although a connection between the Murchison Belt Free State Tonalite + Rubbervale  
356 Volcanics and the Vaalpenskraal Trondhjemite seemed possible based on the age  
357 and Hf isotopic characteristics of the zircons, the zircon  $^{176}\text{Lu}/^{177}\text{Hf}$  ratios (Fig. 3b) are  
358 much higher for the three Murchison Belt samples than for the Makoppa Dome  
359 samples. This is a reflection of the whole rock geochemistry, with flat normalised  
360 REE patterns for the Rubbervale Volcanics (Schwarz-Schampera et al., 2010), and  
361 steeply downward sloping patterns for the Vaalpenskraal Trondhjemite (Anhaeusser  
362 and Poujol, 2004). It therefore seems that the Makoppa Dome samples reflect crustal  
363 reworking, which must have taken place very shortly after juvenile crust formation,  
364 based on the Hf isotopic signature. The Murchison Belt samples, on the other hand,  
365 are evidence of juvenile crust formation, rather than reworking.

366

367 **Kimberley Block**

368 Sample AL3, a fine-grained trondhjemitic dyke from the Amalia area has an age that  
369 is ca. 20 Ma older than the two more K-rich granitoids from the Kraaipan area, MAD1  
370 and KP5, but the three samples are indistinguishable in terms of  $^{176}\text{Hf}/^{177}\text{Hf}$  and  
371  $^{176}\text{Lu}/^{177}\text{Hf}$  (Figure 3a,b). They fall within the same array as the Pietersburg Block  
372 intrusives, with epsilon Hf values of +1 to +2 at ca. 2.9 Ga, and also overlap with  
373 some of the granitoids from the Marydale area described by Cornell et al. (2018). The  
374 one older zircon in sample AL3, at ca. 3.18 Ga, falls slightly off the array for the  
375 Pietersburg Block. It plots very close to the depleted mantle line, and therefore gives  
376 a mantle extraction age of ca. 3.2 Ga, assuming a mafic protolith.

377 Schist sample Madibe-1, presumably a metamorphosed volcanic rock, has an  
378 igneous age of ca. 3.08 Ga, and lies completely off the Pietersburg array, with an  
379 initial epsilon Hf value just above 0. If this isotopic signature reflects reworking of  
380 mafic crust with  $^{176}\text{Lu}/^{177}\text{Hf}$  0.022, then the mantle extraction age is ca. 3.43 Ga  
381 (Table 1); this age would fit with 3.43 Ga zircon found in a volcano-sedimentary  
382 sample from the same area (Poujol et al., 2008). However, if this is part of a TTG  
383 array, with  $^{176}\text{Lu}/^{177}\text{Hf}$  0.0022, then mantle extraction would be around 3.25 Ga. Here,  
384 it must be noted that the scatter of Hf isotope data for this sample is larger than for  
385 the other samples analysed in this study (Fig. 2a), and the data show a rough  
386 negative correlation between  $^{176}\text{Lu}/^{177}\text{Hf}$  ratio and initial  $^{176}\text{Hf}/^{177}\text{Hf}$  ratio (not shown).

387 As zircons can take up HREE during alteration processes (Pidgeon et al., 2019), it is  
388 possible that the spread towards lower  $^{176}\text{Hf}/^{177}\text{Hf}$  ratios is related to this effect. In  
389 that case, the mantle extraction ages quoted here are over-estimates. Either way, the  
390 one older grain of 3.2 Ga that was analysed for its Hf isotopic composition actually  
391 has a  $^{176}\text{Hf}/^{177}\text{Hf}$  ratio that is lower than that of the main population, which is the  
392 opposite of what would be expected if this zircon had been derived from 3.45-3.25  
393 Ga juvenile crust that was the protolith to the Madibe-1 lava. As such, the one older  
394 zircon plots close to the data for the Pietersburg ‘old continental nucleus’ of the  
395 Goudplaats-Hout River gneiss, although a single older zircon may be of limited

396 significance. The main zircon population of sample Madibe-1 shows most similarity in  
397 terms of its Hf isotopic composition and Lu/Hf ratio to samples from the adjacent  
398 Witwatersrand Block (Figure 3a,b), such as the granites studied by Frimmel et al.  
399 (2009) from a borehole west of Carletonville. Considering that this is only a single  
400 sample, it is hard to say whether this can be interpreted as the Madibe greenstone  
401 area having a greater affinity to the Witwatersrand than the Kimberley Block, but,  
402 considering its location so close to the Colesberg Lineament, this possibility cannot  
403 be excluded.

404 Sample BOT1 also plots away from the other Kimberley Block samples, but now on  
405 the young, higher-epsilon Hf side. It shows some similarities in its Hf isotopic  
406 composition to the slightly older Pietersburg Block biotite granite samples GH-2 and  
407 GHBR-2 analysed by Laurent and Zeh (2015). Its signature is however significantly  
408 different from similarly aged granitoids from the Marydale area analysed by Cornell et  
409 al. (2018). The 2.75 Ga age of lapilli tuff sample BOT1, interpreted to be part of the  
410 greenstone basement, was discussed at length by Poujol et al. (2005), since it  
411 appears rather young for the age of the greenstone succession in the area,  
412 especially in the light of the ages of the granitoids of 2.8-3.0 Ga. Some granitoids in  
413 the Marydale area of the Kimberley Block have provided similar ages, which Cornell  
414 et al. (2018) related to crustal melting in connection with plume activity of the  
415 Ventersdorp Supergroup lavas. Although the Ventersdorp volcanism was long  
416 assumed to have started at ca. 2.71 Ga (Armstrong et al., 1991), more recent work  
417 has shown activity at 2791-2779 (Klipriviersberg Group), 2754–2709 (Platberg  
418 Group) and 2709–2683 Ma (Allanridge Group) (Gumsley et al., 2020). Indeed, a  
419 felsic porphyry sample from the Platberg Group of the Ventersdorp Supergroup in the  
420 Amalia area was dated at  $2729 \pm 3$  Ma (U-Pb zircon SHRIMP upper intercept age) by  
421 Poujol et al. (2005). This porphyry was interpreted to overlie the granite-greenstone  
422 succession. It is therefore likely that the lapilli tuffs from which sample BOT1 was  
423 derived are also part of the Ventersdorp Supergroup, rather than the greenstone

424 basement. This ties in with the work by de Kock et al. (2012), who sampled  
425 accretionary lapilli-bearing tuffs near Taung, 50 km to the SW of sample BOT-1, of  
426 the Mohle Formation, which is a local correlative of the Kameeldoorns Formation of  
427 the Platberg Group of the Ventersdorp Supergroup. Zircons from these tuffs gave a  
428 concordia age of  $2735 \pm 3$  Ma. Although no Hf isotope data are available for the  
429 Ventersdorp Supergroup, the Nd isotopic characteristics for the Platberg Group  
430 appear to be subchondritic (see Humbert et al., 2019 for an overview of the existing  
431 isotope data), which does not agree with the decidedly superchondritic Hf isotope  
432 values of the zircon grains of BOT1. Although the Klipriviersberg Group might provide  
433 a match with marginally superchondritic Nd isotope values, the age of  $2754 \pm 5$  Ma  
434 for BOT1 appears to preclude this. More information on the isotopic composition of  
435 the Ventersdorp Supergroup is therefore needed to resolve this matter further. It is,  
436 however, the first evidence of tapping of a juvenile source after ca. 300 Myr of crustal  
437 reworking in the area.

438

#### 439 ***Comparison to detrital zircon data***

440 In as far as our new data overlaps with previously published analyses for granitoids  
441 from the Kaapvaal Craton, they only broaden the choice of provenance areas for  
442 detrital zircon data. However, our data for the Makoppa Vaalskraal trondhjemite  
443 and Amalia lapilli tuff have quite a distinct age-epsilon Hf signature, and can  
444 therefore provide a possible match for detrital zircon grains of which the protosource  
445 has remained elusive.

446 As Figure 4 shows, our three ‘uncommon’ samples (BOT-1, MK10, MK11) only show  
447 limited overlap with published detrital zircon data (see Electronic Supplement 2 for  
448 full references of literature data) from Archean successions, the Magaliesberg  
449 Formation from the Paleoproterozoic Pretoria Group or the Waterberg Group.  
450 However, some overlap is noted between the Vaalskraal trondhjemite samples  
451 MK10 and 11 and detrital zircon from the Eldorado Reef in the upper part of the

452 Witwatersrand Supergroup (Koglin et al., 2010). This is in agreement with  
453 paleocurrent direction from the north and west (Frimmel et al., 2009), and the  
454 connection between the Pietersburg and Witwatersrand blocks (Zeh et al., 2013;  
455 Laurent et al., 2019) around the time of deposition of the upper Witwatersrand  
456 Supergroup at 2.9-2.78 Ga. The characteristics of the accretionary lapilli tuff from the  
457 Amalia area provide a match for zircon from the Svaershoek Formation of the  
458 Waterberg Group (Andersen et al., 2019). It is however doubtful that this equates to  
459 direct derivation of the sediment from these outcrops, considering the depositional  
460 age of ca. 2.05 Ga for the Svaershoek Formation, which is 700 Ma younger than the  
461 Amalia lapilli tuff. This time gap would leave sufficient time for several cycles of  
462 erosion and redeposition to take place. Moreover, if this lapilli tuff indeed belongs to  
463 the Ventersdorp Supergroup rather than the greenstone basement, the potential  
464 protosource area would have had a large areal extent; and, within analytical  
465 uncertainty, the zircon grains from the Waterberg Group also overlap with granitoids  
466 from the Pietersburg Block, so this derivation is ambiguous.

467

#### 468 **Conclusions regarding the evolution of the Kaapvaal Craton**

469 Our new data show that the Makoppa Dome is indeed part of the Pietersburg Block,  
470 despite being separated from the eastern granitoids by the Bushveld Complex, which  
471 itself is obscured by the Paleoproterozoic sedimentary rocks of the Waterberg Group.  
472 The Makoppa Dome samples also provide the first Hf-in-zircon evidence that  
473 reworking of juvenile mafic material started as early as 3.01-3.03 Ga, immediately  
474 after formation of the mafic crust from a depleted mantle source. This is ca. 50 million  
475 years younger than what is known for the eastern Pietersburg Block (Laurent and  
476 Zeh, 2015; Zeh et al., 2013), where the TTGs of the Groot Letaba-Duivelskloof  
477 Domain provide the first evidence for this. This could mean that the tectonic scenario  
478 sketched in Laurent et al. (2019) is actually diachronous, with an earlier onset of  
479 events on the western side of the Pietersburg Block.

480 The data from the three granitoids of the Amalia-Kraaipan area of the Kimberley  
481 block fall within the same epsilon-Hf – age space as Pietersburg granitoids – as do  
482 the granitoids from the Marydale area analysed by Cornell et al. (2018). Therefore,  
483 the Hf isotopic evolution of the two blocks is starting to look remarkably similar,  
484 including evidence for the involvement of older crust (at least as old as 3.2 Ga, based  
485 on near-concordant analyses of older zircon). The schist sample from the Madibe  
486 area could potentially bear evidence of this older crust – although its igneous age is  
487 ca. 3.08 Ga, the Hf isotopic composition of the zircon necessitates a mantle  
488 extraction age of 3.25-3.43 Ga, depending on the assumed protolith. However,  
489 another possibility is that the area, which is very close to the Colesberg Lineament,  
490 bears a closer resemblance to the Witwatersrand Block, for which an older crustal  
491 history is more evident than for either the Kimberley or Pietersburg Block. The  
492 previously reported ca. 2.75 age for a lapilli tuff in the Amalia Belt (Poujol et al.,  
493 2005), which was interpreted as reflecting the age of the greenstone belt, likely  
494 reflects magmatism associated with the Venterdorp Supergroup, although the Hf  
495 isotopic signature of the latter is not known, so direct comparisons cannot be made.

496

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503 and Hugh Rollinson.

504

505

506 **Figure captions:**

507 Figure 1: Overview location map (a), showing boundaries between the blocks after  
508 Eglington and Armstrong (2004); locations for the samples from the Makoppa Dome  
509 (after Anhaeusser and Poujol, 2004) in the Pietersburg Block (b) and from the  
510 Kraaipan-Amalia area (after Poujol et al., 2002) of the Kimberley Block (c). The white  
511 areas in panel (c) are mainly lavas of the Ventersdorp Supergroup. HRSZ = Hout  
512 River Shear Zone; TML: Thabazimbi-Murchison Lineament; PSZ = Palala Shear  
513 Zone; CL= Colesberg Lineament

514

515 Figure 2: Obtained Hf isotope data for the Amalia-Kraaipan (triangles) and Makoppa  
516 Dome (squares) samples in initial  $^{176}\text{Hf}/^{177}\text{Hf}$  (a) and epsilon Hf (b) versus age space.  
517 All data are calculated for the time of igneous zircon crystallization. The inset shows  
518 the data including the younger Archean zircon domains, interpreted to reflect later  
519 reworking or Pb-loss. The typical uncertainties (2 SD) are based on multiple analyses  
520 of reference materials.

521

522 Figure 3: a. Average  $\varepsilon\text{Hf}_i$  versus age for the samples from this study (large symbols),  
523 together with pluton-averages for published analyses, plus single analyses for  
524 inherited (older) zircon grains within the granitoids (references in Electronic  
525 Supplement 2). Trendlines for mafic crust ( $^{176}\text{Lu}/^{177}\text{Hf} = 0.022$ ) and typical TTG  
526 ( $^{176}\text{Lu}/^{177}\text{Hf} = 0.0022$ ) are also shown. The depleted mantle line is following Griffin et  
527 al. (2000). Note that tuff sample BOT1 is more primitive than any other sample, and  
528 likely represents volcanism associated with the Ventersdorp Supergroup. See text for  
529 further discussion.

530 b. Zircon  $^{176}\text{Lu}/^{177}\text{Hf}$  (pluton averages) versus age for the same samples as in a. Note  
531 that the zircons from the Ruberval Volcanics and Free State Tonalite from the  
532 Murchison Belt (Zeh et al., 2013) have much higher  $^{176}\text{Lu}/^{177}\text{Hf}$  than the Makoppa  
533 Dome samples, despite their ages and Hf isotopic compositions being similar. The

534 former are evidence of juvenile crust formation, whereas the latter, together with  
535 slightly younger samples of the Groot Letaba-Duivelskloof (GLD) TTGs (Laurent and  
536 Zeh, 2015) are a result of crustal reworking.

537

538 Figure 4: As Figure 3a, but with the addition of detrital zircon data from Archean  
539 successions, Waterberg Group and Magaliesberg Formation (see Electronic  
540 Supplement 2 for references). Our new Hf isotope data widen the possible  
541 provenance areas for Archean sedimentary rocks, and could explain some of the yet  
542 unmatched grains in the Eldorado Reef of the Witwatersrand Supergroup and  
543 Waterberg Group.

544

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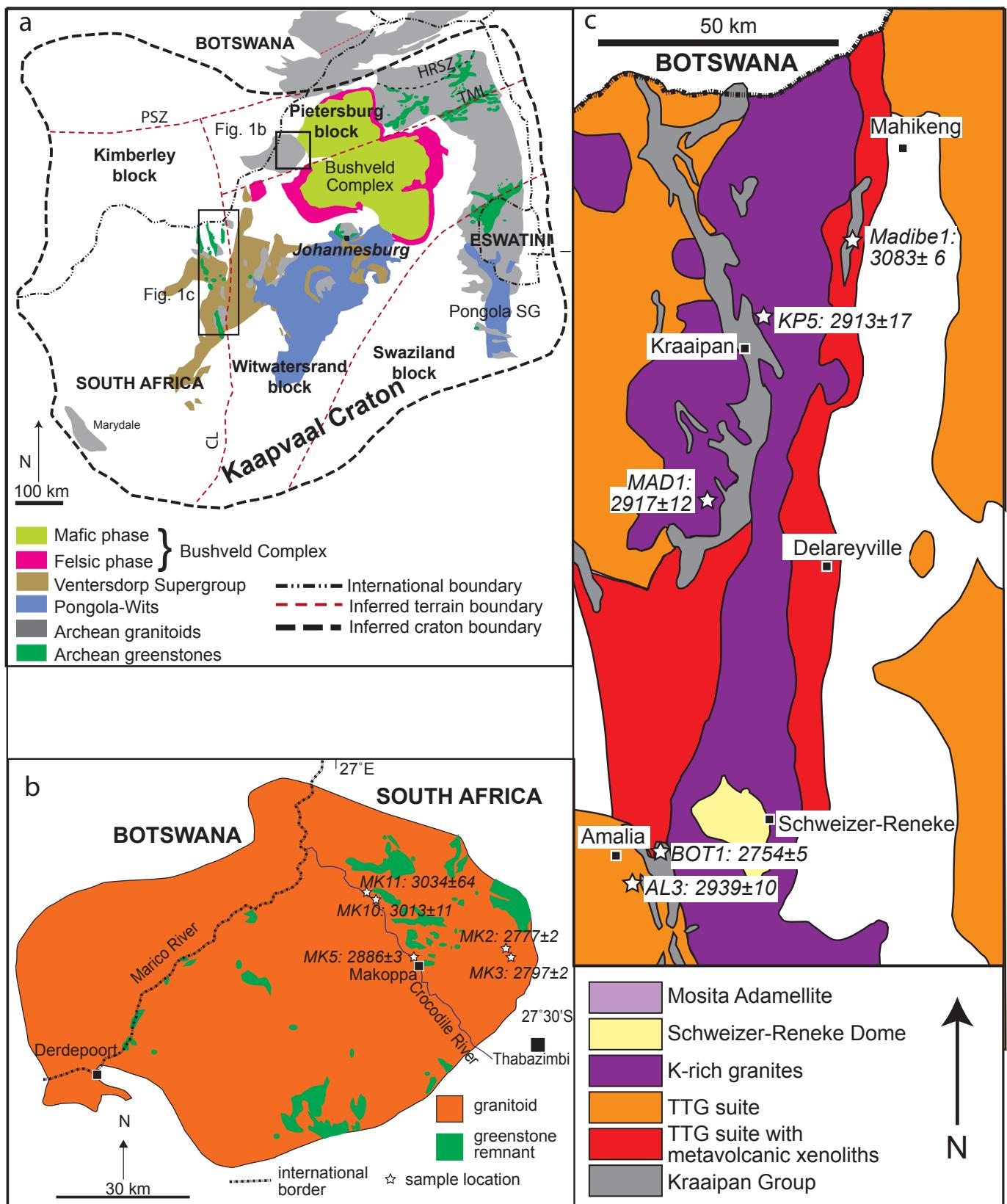


Figure 1 Elburg & Poujol

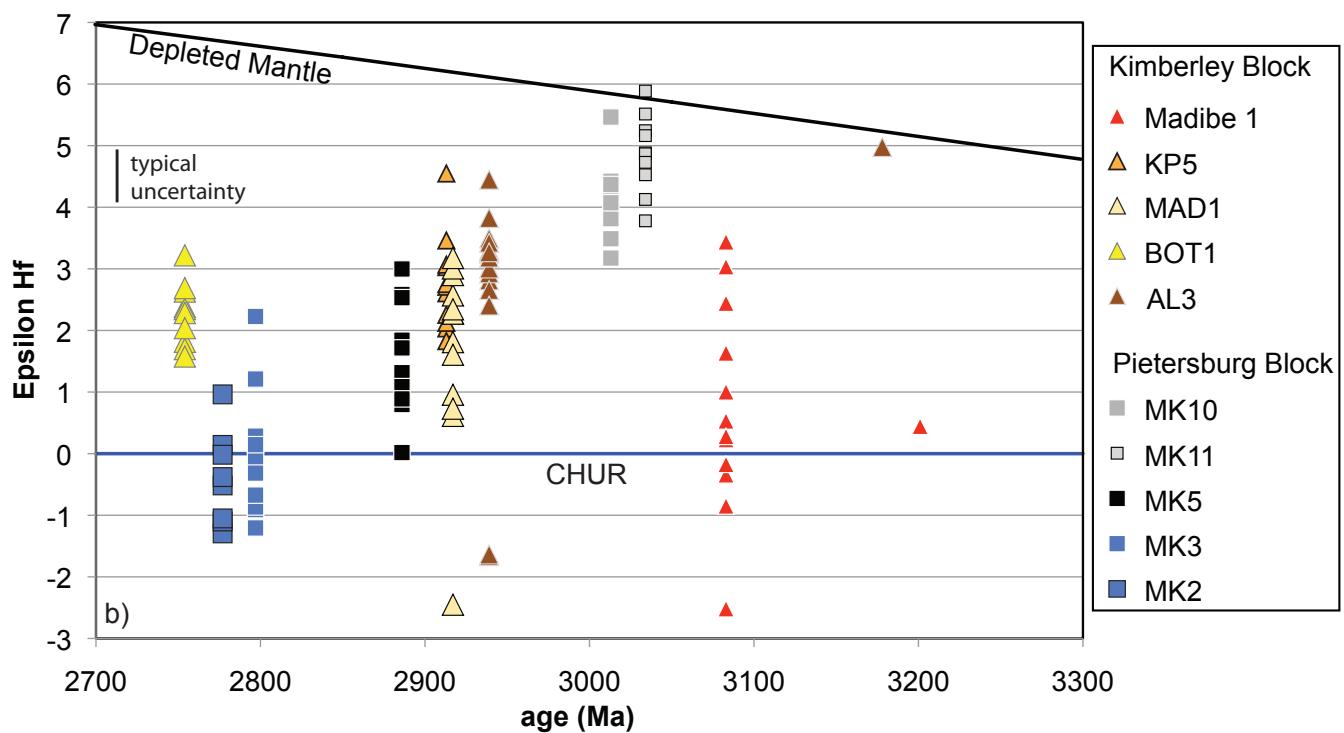
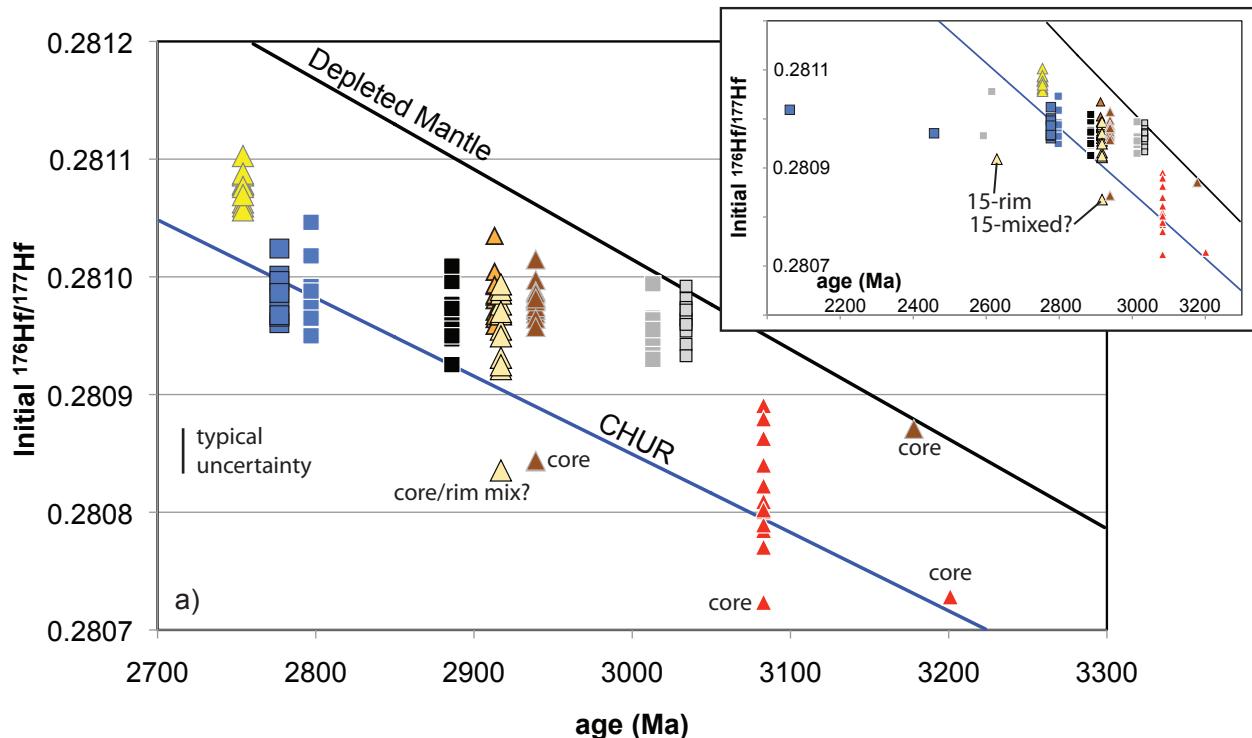


Figure 2 Elburg & Poujol

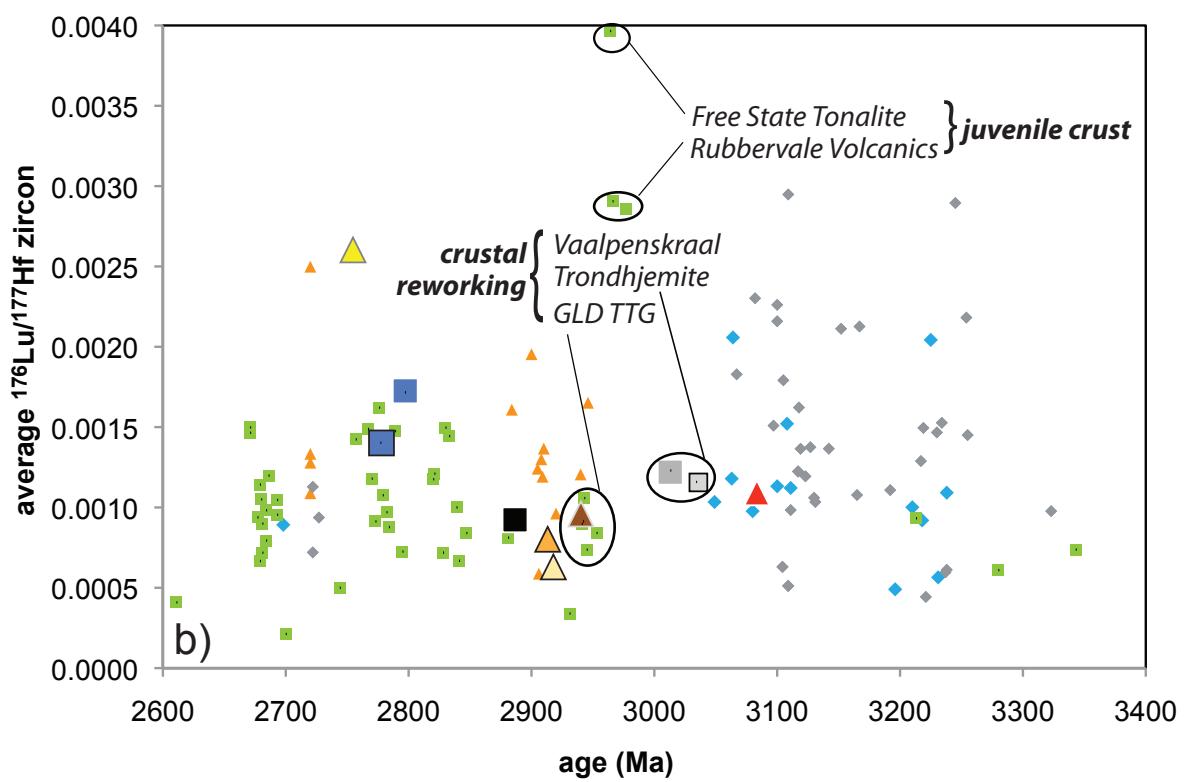
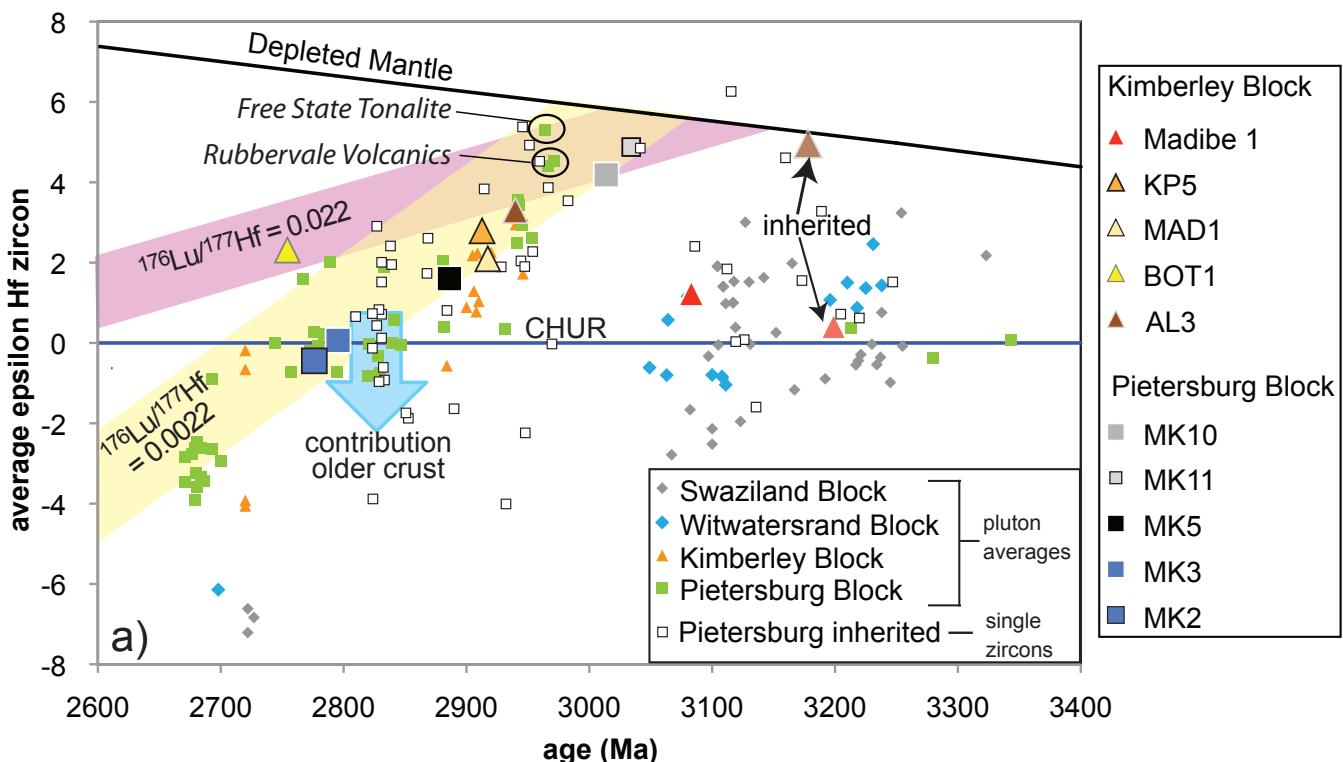


Figure 3 Elburg & Poujol

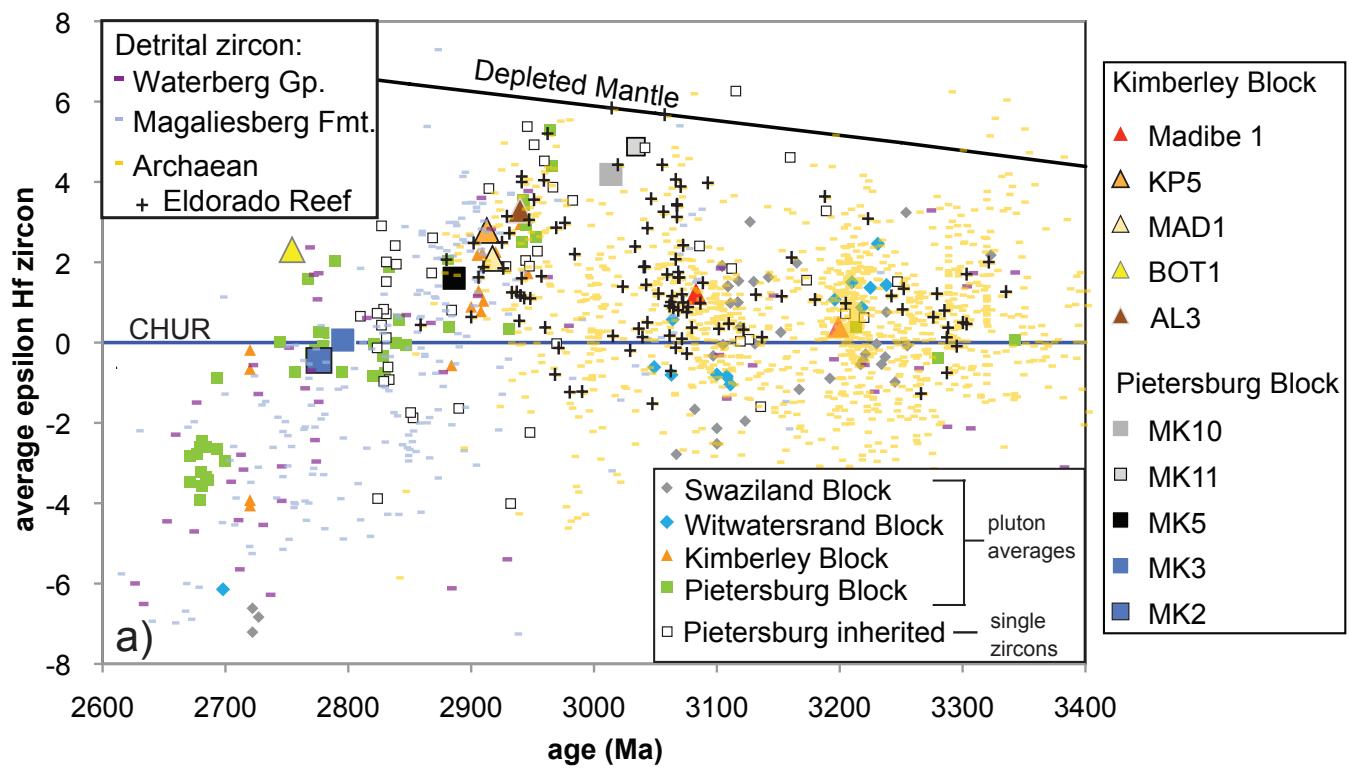


Table 1: Overview of analysed samples.

Sample name	Area	Reference	Rock type	Age ± 2 SD (Ma)	Type of age	Dating technique	mean $^{176}\text{Hf}/^{177}\text{Hf}_\text{i}$	mean $\epsilon\text{Hf}_\text{i}$ (± 2 SD)	$T_{\text{DM}}$ mafic	$T_{\text{DM}}$ tonalitic
<b>Pietersburg Block</b>										
MK10	Makoppa Dome	Anhaeusser and Poujol (2004)	Vaalpenskraal trondhjemite gneiss	3013 ± 11	wtd. av. 7/6	LA-ICP-MS	0.28096	4.2 ± 1.2	3.15	3.08
MK11	Makoppa Dome	Anhaeusser and Poujol (2004)	Vaalpenskraal trondhjemite gneiss	3034 ± 64	wtd. av. 7/6	LA-ICP-MS	0.28096	4.9 ± 1.3	3.10	3.07
MK2	Makoppa Dome	Anhaeusser and Poujol (2004)	Rooibokvlei granodiorite/monzogranite	2777 ± 2	upper intercept	ID-TIMS	0.28099	-0.4 ± 1.5	3.34	3.05
MK3	Makoppa Dome	Anhaeusser and Poujol (2004)	Rooibokvlei granodiorite/monzogranite	2797 ± 2	upper intercept	ID-TIMS	0.28099	0.1 ± 2	3.31	3.05
MK5	Makoppa Dome	Anhaeusser and Poujol (2004)	Makoppa granodiorite/monzogranite	2886 +3/-2	upper intercept	ID-TIMS	0.28097	1.6 ± 1.9	3.26	3.07
<b>Kimberley Block</b>										
MADIBE1	Kraaipan	Poujol et al. (2008)	quartz-sericite schist core	3083 ± 6 3201 ± 4	wtd. av. 7/6 $^{207}\text{Pb}/^{206}\text{Pb}$	SHRIMP SHRIMP	0.28083 0.28073	1.2 ± 3.0 0.4 ± 0.9	3.43	3.25 3.40
BOT1	Amalia	Poujol et al. (2005)	accretionary lapilli tuff	2754 ± 5	wtd. av. 7/6	SHRIMP	0.28108	2.3 ± 1.0	3.11	2.93
MAD1	Kraaipan	Poujol et al. (2002)	grey granodiorite	2917 ± 9	wtd. av. 7/6	SHRIMP	0.28096	2.0 ± 1.8	3.25	3.08
KP5	Kraaipan	Poujol et al. (2002)	pink granodiorite	2913 ± 17	wtd. av. 7/6	SHRIMP	0.28098	2.8 ± 1.5	3.19	3.05
AL3	Amalia	Poujol et al. (2002)	leuco-trondhjemite gneiss core	2939 ± 10 3178 ± 10	wtd. av. 7/6 $^{207}\text{Pb}/^{206}\text{Pb}$	SHRIMP SHRIMP	0.28098 0.28087	3.2 ± 1.0 5.0 ± 1.0	3.22	3.08 3.19

wtd. av. 7/6 = weighted average  $^{207}\text{Pb}/^{206}\text{Pb}$  age $T_{\text{DM}}$  = depleted mantle extraction age in Ga; calculated with  $^{176}\text{Lu}/^{177}\text{Hf} = 0.022$  for mafic protoliths, and 0.0022 for felsic protoliths

The measured  $^{176}\text{Lu}/^{177}\text{Hf}$  ratios for the unknowns were calculated to their initial value using the measured  $^{176}\text{Lu}/^{177}\text{Hf}$  ratios, an  $^{176}\text{Lu}$  decay constant of  $1.687 \times 10^{-11} \text{ year}^{-1}$  (Scherer et al., 2007) and the emplacement ages obtained during previous studies (Table 1). The calculations of epsilon Hf were done with

As shown in Table 1, the measured  $\delta^{17}\text{O}$  values for the chondrites were calculated to a present-day chondritic  $^{17}\text{Ge}/^{77}\text{Ge}$  value of 0.282785 and  $^{17}\text{Lu}/^{17}\text{Lu}$  value of

Analyses in *italics* excluded from average; those underlined exclude  
TDM<sub>2a</sub>: depleted mantle extraction age for mafic protolith

TDM2a: depleted mantle extraction age for tonalitic protolith  
TDM calculated using:

$$\left( \left( \frac{^{37}S_{\text{U}}}{^{37}S_{\text{M}}} \right) \text{DMSO} + \left( \frac{^{37}S_{\text{M}}}{^{37}S_{\text{U}}} \right) \text{DMB} + \left( \frac{^{37}S_{\text{U}}}{^{37}S_{\text{M}}} \right) \text{DTCI} \right) \text{crust.}$$

$$\ln \left( \frac{\left( \frac{L_{\text{IN}}}{L_{\text{IN}} + L_{\text{IR}}} \right) \text{DM,IR} + \left( \frac{L_{\text{IR}}}{L_{\text{IN}} + L_{\text{IR}}} \right) \text{DM,B} + \left( \frac{L_{\text{IN}}}{L_{\text{IN}} + L_{\text{IR}}} \right) \text{crust,I} + \left( \frac{L_{\text{IR}}}{L_{\text{IN}} + L_{\text{IR}}} \right) \text{crust,B} }{ \left( \frac{L_{\text{IN}}}{L_{\text{IN}} + L_{\text{IR}}} \right) \text{DM,B} + \left( \frac{L_{\text{IR}}}{L_{\text{IN}} + L_{\text{IR}}} \right) \text{crust,I} } \right) \approx \lambda$$



Data for figure 3:

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Additional data figure 4:

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