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The multiple ways of recycling Archaean crust: a case study from the ca. 3.1 Ga granitoids from the Barberton Greenstone Belt, South Africa

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1. Introduction

Archaean cratons are stable continental blocks that escaped reworking after their formation (Windley, 1995). They include an Archaean continental crust, underlain by a refractory, rigid lithospheric mantle (Pearson and Wittig, 2014), and they are covered by intracratonic sedimentary successions and possibly cut by dyke swarms (Cawood et al., 2018). The Archaean crust fossilizes early geological evolution, but has typically not been affected by subsequent events: there is a striking contrast between the geological characteristics of the pre-cratonic (Archaean) period and cratonic evolution. Whereas the Archaean regime features soft deformation of a hot crust, a granite-greenstone association, pervasive melting and magmatism, the cratonic stage in contrast records little pervasive deformation or metamorphism. A key issue in Archaean geology is therefore the transition from the Archaean, permobile and hot stage, to the post-Archaean, cold and rigid stage (Capitanio et al., 2019; Chowdhury et al., 2020; Nebel et al., 2018).

In most cratons (Cawood et al., 2018; Laurent et al., 2014a), this transition also corresponds to a change in character of the granitic magmatism, with evolution from pre- to syn-tectonic sodic granitoids of the TTG (tonalite—trondhjemite—granodiorite; Hoffmann et al., 2018; Martin, 1986; Moyen and Martin, 2012) series to a much more diverse assemblage of syn- to post-tectonic, generally potassic granites (Laurent et al., 2014a) formed by a number of processes including differentiation of mantle-derived melts (the “sanukitoid series”) and melting of the crust.

The Kaapvaal Craton of Southern Africa, and in particular the Barberton Granite-Greenstone Terrain in its south-eastern part, offers an excellent example of craton stabilization processes and related changes. Until ca. 3.2 Ga, the geological processes are distinctly “Archaean” in character, with the formation of greenstone belts (Lowe and Byerly, 2007), TTG magmatism (Moyen et al., 2018) and complex – and controversial— deformation and metamorphic patterns (Cutts et al., 2014; Lana et al., 2010; Mühlberg et al., in revision; Schoene and Bowring, 2010; Van Kranendonk et al., 2014; Wang et al., 2019). At ca. 3.07 Ga however, the Dominion Group was deposited over a stable continental basement (Marsh, 2006), forming essentially undeformed sequences unconformably overlying the meso-Archaean basement. The cratonization event in the South-Eastern Kaapvaal craton occurs at ca. 3.1 Ga, and corresponds to a period of emplacement of voluminous granitoids in a relatively low-strain regime (Anhaeusser and Robb, 1983b; Belcher and Kisters, 2006a, b; Westraat et al., 2004). These granitoids formed of granodiorite, monzogranite and syenogranite are collectively referred to as the “GMS” suite (Anhaeusser and Robb, 1983b; Anhaeusser et al., 1983; Anhaeusser et al., 1981). They record the diversity of the craton-stabilizing processes, and are the focus of this work.

In Barberton (as elsewhere), these granitoids are essentially taken as reflecting reworking of the pre-existing TTG-dominated crustal basement (Anhaeusser and Robb, 1983b; Clemens et al., 2010). Their isotopic characteristics (Zeh et al., 2009) as well as those from their counterparts worldwide (Laurent et al., 2014a) are broadly consistent with this model. However, a viable petrological scenario is not as clearly established – partly because of the difficulty to melt large volumes of refractory TTG crust (Clemens et al., 2010; Watkins et al., 2007). Furthermore, the petrological diversity of the GMS suite suggests much more diverse processes.

In this work, we combine a detailed petrological/geochemical interpretation of the composition of the GMS granitoids, with U—Pb and Lu—Hf isotope analyses of samples representative of the different rock types, to propose an integrated model for the formation of the GMS suite and the stabilization of the South-Eastern Kaapvaal Craton at ca. 3.1 Ga. We show that although their petrological diversity requires a range of sources, within and below the crust, and different petrogenetic processes, all the GMS rocks were emplaced in a short period of time at 3110—3105 Ma, and have strikingly similar Hf isotopic characteristics with εHf ca. -2.0 at 3.1 Ga. We discuss the processes permitting this isotopic homogenization, yet preserving petrological diversity. Lastly, we comment on implications for craton stabilizing processes.

2. Geological setting

2.1 The BGGT

The Barberton Granite-Greenstone Terrain (BGGT), and the adjacent Ancient Gneiss Complex of Swaziland, constitute the core of the Kaapvaal craton in Southern Africa, with rocks formed between ca. 3.7 and 3.1 Ga. The Kaapvaal craton is unconformably overlain by several supracrustal sequences mainly formed in intracratonic settings, starting with the Dominion Group (ca. 3.07 Ga), followed by the Witwatersrand (ca. 2.9-2.7 Ga), Ventersdorp (2.73-2.65 Ga), and Transvaal (2.65-2.06 Ga) Supergroups, and further younger sequences. These all attest to the fact that by ca. 3.1 Ga, the Kaapvaal craton had become a stable continental mass, that was intermittently modified by tectono-magmatic processes, leading to the formation of large basins and the Ventersdorp and Bushveld LIP's. The BGGT is made of several units. The Barberton greenstone belt (BGB; review in e.g. Lowe et al., 2012; Lowe and Byerly, 2007) is made up of low-grade supracrustal rocks, starting with mafic/ultramafic sequences of the Onverwacht Group (3530-3260 Ma), followed by the composite Fig Tree Group (3260-3230 Ma) including intermediate to felsic lavas, clastic and chemical (BIF) sediments, and capped by the Moodies Group (3230-3210 Ma) made of a thick sequence of conglomerates and mature sandstones. The Moodies Group unconformably overlies the older units,

and is itself folded in steep, narrow synclines that define today's map pattern (Anhaeusser et al., 1981; Lowe et al., 2012). The change in sedimentation patterns, together with the deformation of the belt, points to a major tectonic event (or events) at Fig Tree to (post) Moodies times, i.e. ca. 3.25-3.20 Ga. This is also the age of amphibolite facies metamorphism, as recorded in the gneiss terrain that surrounds the belt (Cutts et al., 2014; Dziggel et al., 2005; Dziggel et al., 2010; Dziggel et al., 2002; Mühlberg et al., in revision).

The Barberton belt is surrounded by plutonic rocks and orthogneisses. Rocks older than 3.22 Ga are TTGs, and they formed during three main events (review in Moyen et al., 2018) at ca. (i) 3.51 Ga (the Steynsdorp pluton), (ii) 3.45 Ga (the Stolzburg and Theespruit plutons, as well as smaller bodies around them) and (iii) 3.28 Ga (older phases of the Badplaas gneisses) to 3.22 Ga (younger Badplaas gneisses, Nelshoogte and Kaap Valley plutons). The TTG plutons are thus roughly coeval with the formation of parts of the Barberton belt and felsic volcanic units of the same age are recorded in the belt. The felsic schists of the Theespruit Formation emplaced at ca. 3.51-3.53 Ga (Kröner et al., 2016; Kröner et al., 1996; Van Kranendonk et al., 2009), the H6 member of the Hooggenoeg Formation erupted at 3.45 Ga (de Vries et al., 2006; Laurent et al., 2020) and the Fig Tree Group lavas and volcaniclasitic rocks formed at ca. 3.25 Ga (Byerly et al., 1996; Toulkeridis et al., 1999; Zeh et al., 2013).

South of the BGB, the Swaziland Ancient Gneiss Complex (AGC) represents a middle- to lower-crustal domain (Hoffmann and Kröner, 2018; Kröner et al., 2019), presumably exhumed against the BGB at ca. 3.2 Ga along structures now obscured by younger intrusives (Lana et al., 2011; Schoene and Bowring, 2010). The AGC is made of interlayered components including (i) meta-plutonic units, with ages equivalent to the plutonics around the BGB; (ii) meta-sedimentary units with ages corresponding to both the older (Theespruit) components of the BGB (e.g. Kröner et al., 2019; Kröner and Tegtmeyer, 1994; van Schijndel et al., 2017) and the younger (Fig Tree) units (Taylor et al., 2013; Taylor et al., 2016). Collectively, the AGC appears as an equivalent of the BGB and surrounding plutons (or at least of the southern domain of the BGGT, Zeh et al., 2009), buried to mid-crustal depths and deformed, probably during the ca. 3.2 Ga evolution (Kröner et al., 2018; Schoene and Bowring, 2010; Schoene et al., 2008; Taylor et al., 2016).

Plutonic rocks after ca. 3.2 Ga shift to more potassic compositions, ranging from granodiorites, to granites and syenites. They are recorded over a long period of time, from about 3201 Ma for the Dalmein pluton, to less than 2700 Ma for the Mpageni pluton and other 'late granites' of Swaziland (Meyer et al., 1994; Zeh et al., 2009; Zeh et al., 2011). However, they are especially prolific at ca. 3.1

Ga, the emplacement time of the large GMS (granodiorite-monzogranite-syenite) suite that covers large portions of the BGGT (Figure 1).

2.2 The GMS batholiths

The GMS suite consists of four main batholiths (Figure 1): the Nelspruit batholith to the North of the Barberton belt, the Pigg's Peak batholith to the South-East (in Swaziland), the Mpuluzi batholith to the South, and the Heerenveen batholith to the South-West (Anhaeusser and Robb, 1983b; Anhaeusser et al., 1981; Robb et al., 1983). The GMS batholiths form voluminous, sheet-like laccoliths of only a few km thickness, covering large areas, and are composite but dominantly granodioritic to monzogranitic. They are accompanied by smaller bodies of syenite to syenogranite: the Boesmanskop, Weergevonden (also referred as the "Boesmankop tail") and Kees Zyn Doorns intrusions form elongated small bodies emplaced at about the same age as the larger batholiths (Anhaeusser et al., 1983). Existing geochronological constrains (Kamo and Davis, 1994; Westraat et al., 2004; Zeh et al., 2009) point to a short-lived emplacement, between 3110 and 3100 Ma, for the GMS suite.

This study focuses on the Heerenveen batholith, the smallest and best studied of the GMS suite. For comparison, we also include whole-rock data as well as some zircon analyses from the north-eastern margin of the adjacent Mpuluzi batholith.

2.3 The Heerenveen batholith

Located to the southwest of the Barberton Greenstone Belt, the Heerenveen batholith is the smallest of the GMS batholiths, approximately 30×15 km. The Heerenveen batholith has been studied in detail by Belcher and Kisters (2006a, b) who have established the following chronology of emplacement. The Heerenveen batholith is composed of eight different facies emplaced during four main stages of intrusion (Figure 2): (i) Stage I: a first leucogranite (leucogranite I) was emplaced as sheets in the gneissic basement; (ii) Stage II: emplacement of a large homogeneous, weakly deformed, body of porphyritic granite (megacrystic granite), forming the main phase in the Heerenveen (as well as in the Mpuluzi) batholith; (iii) Stage III: a series of smaller, strongly deformed bodies, comprising a second phase of leucogranite (leucogranite II), a quartz-monzonite and a first phase of pink granite (pink granite I), were injected into shear zones developed at the margin of the batholith; and (iv) Stage IV: post-tectonic intrusion of a second phase of undeformed pink granite (pink granite II).

Broadly, this emplacement sequence is also observed in the adjacent portion of the large Mpuluzi batholith [(Moyen et al., in press; Westraat et al., 2004) and unpublished maps and reports by A. Kisters (Stellenbosch University) and students (G. Sonke, C. Campbell, 2006)]]. The Boesmanskop syenite intrusion, now topographically isolated from the Mpuluzi batholith, likely belonged initially to the same intrusive complex. The Heerenveen and Mpuluzi batholiths are separated by a deep valley floored by gneisses, suggesting that the two may have formed an originally continuous carapace (Anhaeusser and Robb, 1983b). On the other hand, the valley separating the two batholiths lies along strike of the Inyoni shear zone, an important feature of the BGGT (Dziggel et al., 2005; Dziggel et al., 2002; Moyen et al., 2006; Van Kranendonk et al., 2014; Wang et al., 2019)(Figure 1). This feature also coincides with an isotopic boundary between two domains, “Barberton North” and “Barberton South” (Zeh et al., 2009), and correlates with a significant geophysical break in the craton (de Wit et al., 1992).

Clemens et al. (2010) gave a detailed account of the diverse facies comprising the Heerenveen batholith, based on the mapping by Belcher et al. (2006a, b) that we also use here (Figure 2). The north-western portion of the Mpuluzi batholith has been described in detail by Westraat et al. (2004) and summarized below (Figure 3). All the facies encountered in the Heerenveen batholith contain quartz, microcline and plagioclase, in various proportions. The porphyritic crystals in the megacrystic granite are mostly euhedral microcline, or in some outcrops quartz. Muscovite has been observed in sheared leucogranites I & II. Biotite, typically chloritized, is the ubiquitous mafic mineral, and the quartz-monzonite also contains hornblende and euhedral titanite.

This work is based on whole rock geochemistry of the Heerenveen (and parts of Mpuluzi) batholiths, as well as U-Pb dating and Hf isotope analyses of zircons from selected samples. Fourteen samples from Heerenveen and four from Mpuluzi have been studied. They cover most of the lithologic and geochemical diversity of the batholiths.

2.4 The Lower Onverwacht Group

Meta-sediments have been suggested as a possible source for the Heerenveen magmas (Clemens et al., 2010), and were consequently investigated in this work.

The lowermost portion of the Onverwacht Group mostly comprises mafic and ultramafic schists and amphibolites, but also contains some layers of felsic schists that are regarded as felsic meta-volcanic or volcaniclastic rocks (Anhaeusser et al., 1981; Kröner et al., 1996; Lowe et al., 2012; Lowe and Byerly, 2007; Van Kranendonk et al., 2009). Felsic schists occur in two formations, the Sandspruit and Theespruit Formations. The Theespruit formation is relatively coherent, whereas most of the Sandspruit Formation occurs as enclaves in the plutons and gneisses of the Stolzburg block South of

the BGB. As formation-level correlations are difficult, and beyond the scope of this work, we do not attempt further subdivision and rather refer to the Lower Onverwacht Group.

The felsic schists were dated in two areas, around the Steynsdorp pluton (Kröner et al., 2016; Kröner et al., 1996) and North of the Theespruit pluton, in the footwall of the Komati fault (e.g. the "Tjakastad water tower" locality; Kröner et al., 2016; Van Kranendonk et al., 2009). In both areas, the age of the protolith is ca. 3530 Ma. The Lower Onverwacht Group also includes rare occurrence (reported in Dziggel et al., 2002) of a coarse-grained quartz-plagioclase-microcline-clinopyroxene clastic meta-sediment with a maximum age of sedimentation of ca. 3520 Ma, and interpreted as a meta-arkose.

In order to further characterize the Lower Onverwacht Group, and explore its suitability as a source for the GMS rocks, we included five samples of it in our study. Four samples are felsic agglomerates from the Tjakastad Schist Belt (Diener et al., 2005), a previously undated portion of the Lower Onverwacht Group. The fifth sample comes from an 'arkose' fragment, sample (BE5), previously studied by Dziggel et al. (2002) and Diener and Dziggel (in press).

3. Analytical methods

3.1 Bulk rock composition

Bulk rock compositions for samples collected in this work were commercially obtained at ALS Global labs Ireland, using their "Complete Characterization Package" (see details at https://www.alsglobal.com/en-au/services-and-products/geochemistry/geochemistry-testing-and-analysis/whole-rock-analysis-and-lithogeochemistry). In addition, we compiled a database of published rock compositions drawing primarily on data by Anhaeusser et al. (1983a), Westraat et al (2004) and Clemens et al. (2010). Existing analyses do not always report trace elements and the final database (Data Repository, Item DR1) includes 88 new analyses, mostly of the Heerenveen batholith, and 289 from the literature. In total, we have 28 analyses from the Boesmanskop, 197 from the Mpuluzi, and 152 from the Heerenveen intrusions. Most of the published data does not include reference to the facies that we describe here and accordingly have been assigned on the basis of (i) the field description, when available; (ii) geochemical properties ; and (iii) the geographic location.

3.2 U-Th-Pb isotope analyses

Uranium, thorium and lead isotopes were analyzed using a ThermoScientific Element 2 sector field ICP-MS coupled to a Resolution M-50 (Resonetics) 193 nm ArF excimer laser (ComPexPro 102F, Coherent) system at Goethe-University Frankfurt (GUF). Data were acquired in time resolved – peak

jumping – pulse counting / analogue mode over 466 mass scans, with a 20 second background measurement followed by 21 second sample ablation. Laser spot-size was 19 µm for unknowns and the standard zircon OG1, 33 µm for the standard zircons GJ1 (primary standard), and 40 µm for the standard zircon 91500. The results of the standard zircon measurements obtained during the different analytical sessions are shown in Data Repository, item DR2. Sample surface was cleaned directly before each analysis by three pulses pre-ablation. Ablations were performed in a 0.6 l min^{-1} He stream, which was mixed directly after the ablation cell with 0.007 l min^{-1} N_2 and 0.83 l min^{-1} Ar prior to introduction into the Ar plasma of the SF-ICP-MS. All gases had a purity of >99.999% and no homogeniser was used while mixing the gases to prevent smoothing of the signal. Signal was tuned for maximum sensitivity for Pb and U while keeping oxide production, monitored as $^{254}UO/^{238}U$, below 0.7-0.3%. The sensitivity achieved was in the range of 9000-14000 cps/µg g^{-1} for ^{238}U with a 26 µm spot size, at 5.5 Hz and about 4 J cm^{-2} laser energy. The typical penetration depth was about 10-20 µm (spot size dependent). The two-volume ablation cell (Laurin Technic, Australia) of the M50 enables detection and sequential sampling of heterogeneous grains (e.g., growth zones) during time resolved data acquisition, due to its quick response time of <1s (time until maximum signal strength was achieved) and wash-out (< 99.9% of previous signal) time of about 2s. With a depth penetration of ~0.7 µm.s^{-1} and a 0.46 s integration time (4 mass scans = 0.46 s = 1 integration) any significant variation of the $^{207}Pb/^{206}Pb$ and $^{238}U/^{206}Pb$ in the µm scale is detectable. Raw data were corrected offline for background signal, common Pb, laser induced elemental fractionation, instrumental mass discrimination, and time-dependent elemental fractionation of Pb/U using an in-house MS Excel© spreadsheet program (Gerdes and Zeh, 2006; Gerdes and Zeh, 2009). A common-Pb correction based on the interference- and background-corrected ^{204}Pb signal and a model Pb composition (Stacey and Kramers, 1975) was carried out. The ^{204}Pb content for each ratio was estimated by subtracting the average mass 204 signal, obtained during the 20 second baseline acquisition, which mostly results from ^{204}Hg in the carrier gas (c. 230-350 cps), from the mass 204 signal of the respective ratio. For the analyzed sample the calculated common ^{206}Pb contents was mostly <0.5% of the total ^{206}Pb but in very rare cases exceeded 4% (Data Repository, item DR2). Laser-induced elemental fractionation and instrumental mass discrimination were corrected by normalization to the reference zircon GJ-1 (Jackson et al., 2004). Prior to this normalization, the inter-elemental fractionation ($^{206}Pb^*/^{238}U$) during the 21s of sample ablation was corrected for each individual analysis. The correction was done by applying a linear regression through all measured, common Pb corrected ratios, excluding the outliers (± 2 standard deviation; 2SD), and using the intercept with the y-axis as the initial ratio. The total offset of the measured drift-corrected $^{206}Pb^*/^{238}U$ ratio from the "true" ID-TIMS value (0.0983 ± 0.0004; ID-TIMS GUF value) of the analyzed GJ-1 grain was between 4 and 14% (during all analytical sessions) and the drift over the day, during the different analytical session between 1 and 8%.

Reported uncertainties (2σ) of the $^{206}Pb/^{238}U$ ratio were propagated by quadratic addition of the external reproducibility (2 SD) obtained from the standard zircon GJ-1 (see Data Repository, item DR2) and the within-run precision of each analysis (2 SE; standard error). In case of the $^{207}Pb/^{206}Pb$ we used a ^{207}Pb signal dependent uncertainty propagation (Gerdes and Zeh, 2009). The $^{207}Pb/^{235}U$ ratio is derived from the normalized and error propagated $^{207}Pb/^{206}Pb^*$ and $^{206}Pb^*/^{238}U$ ratios assuming a $^{238}U/^{235}U$ natural abundance ratio of 137.88 and the uncertainty derived by quadratic addition of the propagated uncertainties of both ratios. The analytical results are presented in data repository. The accuracy of the method was verified during the different sessions by analyses of reference zircon 91500, Plešovice, and OG1, that yielded Concordia ages of 1065.5 ± 6.7 (n=7, MSWD=0.22, Prob.=0.89); 339.1 ± 2.4 Ma (n=32; MSWD=0.98, Prob.=0.32), and 3461.3 ± 6.4 Ma (n=45; MSWD=0.77, Prob.=0.38), respectively (Data Repository, item DR2), identical within error of the quoted TIMS values of 1065.4 ± 0.4 Ma (Wiedenbeck et al., 2004), 337 ± 0.37 Ma (Sláma et al., 2008), and of 3465.4 ± 0.6 Ma (Stern et al., 2009). The data were plotted using ISOPLOT (Ludwig, 2001)

3.3 Lu-Hf isotope analyses

Hafnium isotope measurements were performed with a Thermo-Finnigan NEPTUNE multi collector ICP-MS at GUF coupled to the same laser as described in the U-Pb method (Resolution M-50 193 nm ArF excimer laser). Round or rectangular laser spots with diameters or edge lengths of 33-40 µm were drilled with repetition rate of 5.5 Hz and an energy density of about 6-7 J/cm^2 during 55 s of data acquisition. All data were adjusted relative to the JMC475 of $^{176}Hf/^{177}Hf$ ratio = 0.282160. Quoted uncertainties are quadratic additions of the within run precision of each analysis and the reproducibility of the zircon standard GJ1 which ranged between 0.0048% and 0.0070% (2SD) during the different analytical sessions (Data Repository, item DR3). Accuracy and external reproducibility of the method was verified by repeated analyses of reference zircons GJ-1, 91500, and Plešovice, which yielded $^{176}Hf/^{177}Hf$ of 0.282003 ± 0.000024 (2SD, n=9), 282290 ± 0.000022 (n = 9), and of 0.282465 ± 0.000019 (n=7), respectively (Data Repository, item DR3). These ratios are well within the range of solution mode data (Gerdes and Zeh, 2006; Woodhead and Hergt, 2005).

For calculation of the epsilon Hf [εHf_t] the chondritic uniform reservoir (CHUR) was used as recommended by Bouvier et al. (2008), i.e. $^{176}Lu/^{177}Hf$ and $^{176}Hf/^{177}Hf$ of 0.0336 and 0.282785, respectively, and a decay constant of 1.867 x10^{-11} (Scherer et al., 2001; Söderlund et al., 2004). Initial $^{176}Hf/^{177}Hf_t$ and εHf_t for all analysed zircon domains were calculated using the respective intrusion ages (Data Repository, item DR3). Depleted mantle hafnium model ages (T_{DM}) were calculated using values for the depleted mantle as suggested by Blichert-Toft et al. (2010) , with $^{176}Hf/^{177}Hf$ =

0.283294 and a $^{176}Lu/^{177}Hf$ of 0.03933, corresponding to a straight DM-evolution line with εHf_{today}=+18 and $\varepsilon Hf_{4.558Ga}$=0. T_{DM} ages for all data were calculated by using the measured $^{176}Lu/^{177}Hf$ of each spot for the time since zircon crystallization (= intrusion age), and a mean $^{176}Lu/^{177}Hf$ of 0.0113 for average continental crust (average from Taylor and McLennan, 1985; Wedepohl, 1995).

4. Geochemistry of the Heereveen batholith

From a geochemical perspective, the multiple facies of the GMS rocks in the southern BGGT are divided into three main groups and several sub-groups (Figure 4; Table 1). The first two groups (low-Ca granitoids, and syenites and related rocks) match the descriptions of Clemens et al. (2010), who refer to these as high and low SiO_2 respectively (limit at 71 %). The high-SiO_2 group (our low-Ca granitoids) is the dominant rock type, and only some of the stage III intrusives (monzonites, pink granite I) belong to the low SiO_2 group (our syenites). We identify here, in addition, a group of "dark granitoids" mostly present in the Mpuluzi batholith but also forming rare dykes in the Heerenveen body. They are petrographically similar to the Hebron phase of the Nelspruit Batholith (Cornwright et al., 2008).

4.1 Major elements and granite types

Most chemical classifications of granitoids (Bonin et al., 2020; Debon and Lefort, 1983; Frost et al., 2001; Moyen, 2019; Stussi, 1989) acknowledge that a handful of parameters are required for a full chemical description, and these parameters appear in different forms in commonly used diagrams (Figure 4) : (i) indicators of *differentiation* (e.g., SiO_2, or FeOt + MgO); (ii) proxies for the *plagioclase to K-feldspar* ratio, e.g. molar balance between Ca, Na and K ; (iii) the balance between *Na and K*; (iv) estimates of the *excess or deficit* of *Al* relative to the feldspar stoichiometry (A/CNK, normative corundum, etc.); (v) the balance between *Fe and Mg* (mg#, FeOt/MgO, etc.). The GMS granitoids are here described in these terms.

4.1.1 The dominant low-Ca granitoid types

The term "low Ca" was initially proposed by Champion et al. (1997) to describe large, leucocratic batholiths, generally syn-tectonic and shallowly emplaced, in the Yilgarn craton of Western Australia. The Yilgarn low-Ca granitoids have variable K/Na ratios, and are associated with minor units of more mafic granitoids, emphasizing their similarity with the Barberton GMS suite.

The GMS low Ca granitoids are leucocratic (SiO_2 > 71 %) and are represented by the main megacrystic phases of both the Mpuluzi and Heerenveen batholiths, as well as the leucogranites in the two bodies. They constitute the bulk, perhaps 90 %, of the Heerenveen and Mpuluzi batholiths and collectively represent large volumes of magma. They are weakly peraluminous, although for such

SiO_2 levels this is not a discriminatory feature as most granites converge towards this portion of geochemical space (Bonin et al., 2020). They are calc-alkalic to alkali-calcic and may be magnesian or ferroan, although this too is non-diagnostic. They also range from low to high K_2O (1 to 5 %), and can on this basis be subdivided into three sub-groups (low, medium and high K_2O; Table 1), that correspond to different mappable facies (Table 1; Figure 2). The high-K_2O subgroup tends to be more aluminous than the other subgroups.

In general, the low-Ca granitoids of the Mpuluzi batholith are more potassic than their counterparts in the Heerenveen batholith. The main Mpuluzi megacrystic granite is high-K_2O, and so are the majority of the leucogranites. One facies only has intermediate K_2O contents. In contrast, the main Heerenveen megacrystic granite and the leucogranite I are low in potassium, and only the Heerenveen leucogranite II has intermediate K_2O contents.

4.1.2 The late syenites and related rocks

Syenites, monzonites and pink granites represent a minor (5-10 %) component of the Heerenveen and the Mpuluzi batholiths (Figure 2), as well as the bulk of the Boesmanskop intrusion. They define a magmatic series, ranging from ca. 60 to 70 % SiO_2, and are invariably high- to ultra-high-K_2O (5 – 6 % K_2O even at lower SiO_2 contents). They are mostly metaluminous, ferroan and alkali-calcic to alkalic. At high SiO_2, the composition of the syenites overlaps with the high-K_2O members of the low Ca group, but they belong to distinct mappable units and thus represent different magmas.

4.1.3 Minor dark granites

Dark granites constitute a rather heterogenous, volumetrically small, group of lithologies (perhaps 1-5 %), seldom large enough to be discriminated on a map. They are generally late in the intrusion sequence, forming small dykes in older lithologies. Along the edge of the Mpuluzi batholith (Figure 3g), they occur as the matrix of a spectacular intrusion breccia with clasts of syenogranites wrapped in a dark augen granodiorite. In the core of the Mpuluzi batholith, published databases (unfortunately often lacking facies or outcrop descriptions: Anhaeusser and Robb, 1983a; Murphy, 2015) reveal the presence of scattered occurrences of chemically similar rocks, perhaps corresponding to the “veins” mapped in portions of the Mpuluzi batholith by Anhaeusser et al. (1981). In the Nelspruit Batholith, similar rocks form a discrete intrusion, the Hebron pluton (Anhaeusser et al., 1981; Cornwright et al., 2008). All of this is suggestive of late, syn-plutonic dykes injected into the partially consolidated magma reservoir, and possibly undergoing fragmentation into microgranular mafic enclaves (Barbarin, 2005).

The dark granitoids mostly have 60-65 % SiO_2, and variable characteristics summarized in Table 1. They define several series ranging from a low-K_2O, slightly peraluminous series to a high-K_2O,

metaluminous series, and in most diagrams (Figure 4) the various rock types occupy positions intermediate between the syenites and the other groups. Here too, the most differentiated units overlap in composition with the low-Ca granites, but the field context demonstrates that they belong to different magma batches.

4.2 Trace elements

4.2.1 An improved representation of trace element trends in granitoids

Trace elements are, in general, not particularly good petrogenetic tracers in granitoids (Bonin et al., 2020; Janoušek et al., 2016; Moyen and Laurent, 2018). Distributions are mostly controlled by accessory minerals (e.g. LREE in allanite or monazite, Zr and Hf in zircon, etc.), such that they provide little information on sources and processes. Like all igneous rocks, the composition of granitic melts is a function of the source composition and the melting process. Unlike mantle-derived rocks, however, the magnitude of the differences between different sources is much more important in granitoids than it is in mafic rocks, and therefore the trace element composition of granitoids is (with a few exceptions, such as the HREE fractionation in presence of garnet and amphibole) primarily a reflection of their source (Janoušek et al., 2010; Konopásek et al., 2018).

One may take advantage of this by devising spidergrams that magnify the differences between potential sources, thereby revealing meaningful petrogenetic processes.

In the original construction of spidergrams, elements are normalized against the inferred source, in this case the mantle, and arranged in order of decreasing incompatibility relative to MORB (Coryell et al., 1963; Masuda, 1957, 1962). Any deviation from a smooth pattern (or “anomaly”) can be interpreted as reflecting the effects of specific minerals – for instance Fe-Ti-oxides on the abundances of Nb and Ta. Following the same logic for granites, it is therefore useful to normalize the studied sample against a potential source, here the average Barberton TTGs (Table 2; Moyen et al., 2018), and Figure 5 depicts REE diagrams normalized to the average of Barberton TTG. Chondrite-normalized REE diagrams, for comparison, are in Figure SE1: supplementary online material. To further mimic the MORB approach, elements should be arranged in order of decreasing incompatibility with respect to a crustal melting scenario. In Figure 6 we show spidergrams that are (i) normalized to average TTGs and (ii) arranged by increasing variability: Schiano et al. (1993) suggested that variability within a suite correlates to bulk distribution coefficients, thus increasing variability should correspond to increasing compatibility.

This representation allows to evidence two important features. Firstly, the trace elements patterns within each group (or subgroup) are similar, confirming that each group forms a magmatic series in

which rock compositions are related by simple processes. Secondly, the groups show different patterns. Therefore, they can hardly relate to the evolution of a common parental magma. A key observation is thus the presence of a very large range of petrologically unrelated magmas, as detailed below.

4.2.2 The low-Ca granitoids

The three sub-groups of the low-Ca granitoids are broadly similar, but with consistent differences. The low-K_2O sub-group has trace element patterns that are flat and close to normalized values of 1 and, therefore, do not differ much from an average Barberton TTG. The high-K_2O sub-group is up to 10 times enriched, and shows prominent anomalies for Eu, Sr and Ti. The medium K_2O sub-group exhibits intermediate characteristics.

The overall flat shape of the patterns is consistent with melting of a source with TTG-like trace elements composition. Other Archaean felsic rocks may however have similar trace element signatures, for instance clastic sediments derived from TTG erosion or composite Grey Gneisses containing a significant (meta-)TTG component. Sr, Eu and Ti anomalies suggest that the (high-K) melts coexisted with plagioclase and Ti-oxides at some point of the magma's evolution – either as a residual phase during melting, or a fractionated phase during crystallization.

In either case, the 10-fold enrichment observed in the high-K_2O sub-group suggests melt fractions of ca 0.1. With decreasing concentration of incompatible elements, and thus higher melt fractions, the anomalies are less pronounced, consistent with the progressive separation of melt and solid during melting or crystallization..

4.2.3 The syenites and dark granites

Both types of dark granitoids, as well as the syenites, have normalized REE patterns that are steeper than those of the low-Ca rocks. They have a small or insignificant Eu anomaly, and the typical REE pattern is S-shaped. Such a shape seems at odds with an origin from a TTG-like source (as suggested for the low-Ca granitoids), as this would require unprecedented decoupling of the REEs. Similarly, for the extended trace element patterns of Figure 6, the increase towards more compatible elements would imply, for rocks with a TTG-like source, a reversed order of incompatibility which is unlikely.

It is therefore much more likely that these rocks originated from a different source (see later for a more detailed discussion). Compared to TTGs, this source would have had negatively-sloped TTG normalized REE patterns. In a more conventional mantle- or chondrite-normalized diagram, the corresponding patterns would be steeper than TTGs. In terms of other trace elements, the source was TTG-like and more enriched in HFSE and HREE than in LILE and LREE. Negative anomalies in Sr or

Ti may either correspond to residual minerals (as was the case for the low Ca granitoids), or reflect such anomalies already in the source.

5. U—Pb—dating and Lu-Hf isotope results

5.1. The GMS samples

All 18 samples of the GMS suite yield magmatic zircon populations. Grains are euhedral and range from elongate, needle-like crystal habits (aspect ratios up to 9), to stubby morphologies (aspect ratios 2 to 3), even within individual samples (Figure 7). Zircons in the samples from the Mpuluzi batholith are commonly much larger and more equant (average length = 195 µm, width = 82 µm, aspect ratio = 2.5, n=185) compared to those from the Heerenveen samples (average length = 162 µm, width =52 µm, aspect ratio =3.4, n=506). In all samples zircons show an oscillatory zoning, but many grains also reveal altered domains (recrystallized and porous), or are highly fractured. Optically distinct cores are rare, and mainly were found in sample 11-15B, where CL-bright cores are commonly surrounded by CL-dark, strongly altered rims (Figure 7). Th/U ratios of all measured zircons ranges from 0.5 to 2, consistent with what is commonly observed in igneous zircons.

Example Concordia diagrams for the studied samples are presented in Figure 8, with the other diagrams in Figure SE2: supplementary online material. Zircons of most samples yielded similar Concordia ages at around 3105 Ma, with the most reliable dates (comprising at least 3 concordant analyses per samples) ranging from 3096 ± 19 to 3114 ± 12 Ma. These ages are in agreement with upper intercept ages obtained from the same samples, and with U-Pb ages previously obtained by ID-TIMS dating (Kamo and Davis, 1994; Westraat et al., 2004; for comparison see Table 3). Significantly older $^{207}Pb/^{206}Pb$ ages at about 3200 Ga were only found in 4 samples from the Heerenveen batholith, where eight analyses of zircon cores of sample HE 11-15B (in Figure 1: supplementary online material) yielded a Concordia age of 3201 ± 9 Ma. This age is slightly younger than that of the youngest TTG's observed in the Barberton Greenstone belt, which were emplaced at 3230-3210 Ma (see compilation in Moyen et al., 2018).

Hafnium isotope analyses obtained from zircon populations of all GMS samples, independent of composition and location are all similar (mostly identical within error) as shown in Table 4. Initial $^{176}Hf/^{177}Hf$ ratios range between 0.28071 ± 0.00003 and 0.28076 ± 0.00002 (2 sigma standard deviations), corresponding to εHf_t values between -0.9 ± 0.7 and -2.5 ± 1.7, and Hf model ages between 3.48 ± 0.02 Ga and 3.57 ± 0.05 Ga, respectively. Identical initial $^{176}Hf/^{177}Hf$ ratios of concordant and discordant zircons in individual samples suggest that all these grains/domains crystallised from the same isotopically homogeneous magma, but that certain domains underwent

significant Pb loss due to alteration, that nevertheless left the Hf isotope composition unchanged (Figure 2: supplementary online material; for more explanations see Gerdes and Zeh, 2009). Identical initial $^{176}Hf/^{177}Hf$ ratios of inherited zircons in sample HE 11-15B and magmatic zircons in the GMS indicate that magmas forming at c. 3.2 Ga and c. 3.1 Ga in the BGB all had similar Hf isotopic compositions. The Hf isotopic compositions of the GMS zircons overlap with those of the 3.23 Ga TTG's of the Barberton North domain (Kaap Valley and Nelshooghte), showing $^{176}Hf/^{177}Hf$ ratios between 0.28073 ± 0.00002 and 0.28076 ± 0.00002, respectively (Zeh et al., 2009).

5.2. The Lower Onverwacht metasediments

Five samples from the Lower Onverwacht Group have been analysed. Four samples are meta-volcanic or volcaniclastic 'agglomerates' from the Tjakastad Schist Belt (Diener et al., 2005). The other is a clastic meta-sediment ('arkose') from a greenstone remnant in the southern portion of the Stolzburg terrane (Dziggel et al., 2002).

Zircon grains of the four agglomerate samples are angular, have oscillatory and/or sector zoning, and contain recrystallized melt inclusions and apatite. Many grains show signs of alteration, re-crystallization and intense fracturing (Figure 7). Zircon analyses yield concordant and discordant ages, with the oldest domains in all four samples plotting on Discordia lines with upper intercept ages around 3530 Ma. The most pristine zircon domains yield Concordia ages between 3524 ± 7 Ma and 3531 ± 7 Ma (Table 3; Figure 8e; Figure SE2: supplementary material). These ages overlap with those for Theespruit Formation meta-volcanics in the nearby Tjakastad area (ca. 3530 Ma; Kröner et al., 2016; Van Kranendonk et al., 2009). Younger ages, perhaps result from subsequent Pb-loss, either related to emplacement of the adjacent Theespruit and Stolzburg Plutons at 3450 Ma, or later tectono-metamorphic-magmatic events which affected the BGGT at 3.23 Ga (Cutts et al., 2014; Diener et al., 2005; Diener and Dziggel, in press; Heubeck et al., 2013; Schoene and Bowring, 2010). Hafnium isotope analyses of zircon populations from the four agglomerates yielded similar initial $^{176}Hf/^{177}Hf$ ratios. These range between 0.28046 ± 0.00006 and 0.28048 ± 0.00007 (2 S.D.), corresponding to εHf_t values between -1.4 ± 2.2 and -0.6 ± 2.4, and Hf model ages between 3.80 ± 0.13 Ga and 3.83 ± 0.12 Ga, respectively. The relatively wide scatter in εHf_t values (more than 4 epsilon units) of individual samples, suggests that the Theespruit meta-volcanics were sourced from magmas of variable composition, perhaps resulting from different crust and mantle contributions. The meta-agglomerate data overlap with those obtained by Kröner et al. (2016) from Theespruit Formation felsic tuffs of the nearby Tjakastad area, that show an even wider range in εHf_t values from – 6.2 to +0.5.

Forty-five zircon grains with magmatic oscillatory zoning, and minor signs of alteration were analysed from the meta-arkose. Eighteen grains yielded concordant ages between 3510 and 3450 Ma, of which nine analyses yielded a Concordia age of 3510 ± 8 Ma [MSWD(c.+e.)=0.70, Probability(c.+e.)=0.80, c.+e. = concordance and equivalence] and 4 analyses 3464 ± 8 Ma [MSWD(c.+e.)=0.30, Probability(c.+e.)=0.95]. Two analyses gave concordant ages at about 3200 Ma (Figure 8f). The concordant age at 3510 Ma is slightly younger than the deposition age of 3520 Ma proposed by Dziggel et al. (2005), and also younger than the emplacement ages of ca. 3530 Ma for the agglomerates of the Tjakastad Schist belt (this study), and of 3534.4 ± 1.5 Ma, reported by Kröner et al. (2016) for the felsic tuffs from the same locality. The concordant age at c. 3465 Ma is slightly older than the emplacement age of the enclosing Stolzburg trondhjemites. Ages of ca. 3.2 Ga overlap with those reported by Wang et al. (2019) from zircons found in the immediate vicinity (samples 14SA36, 15SA36), suggesting some resetting of the ages during younger tectono-metamorphic events. Initial $^{176}Hf/^{177}Hf$ ratios of the detrital zircons in Theespruit arkoses (n=33) are generally higher than those obtained from the agglomerates, and reveal two clusters: (1) at $^{176}Hf/^{177}Hf$ = 0.280572 ± 0.000024 (n=18), corresponding to $\varepsilon Hf_{3.45\text{-}3.51Ga}$ =2.0 ± 0.7, and T_{DM}=3.66 ± 0.05 (4 zircons with ages between 3.45-3.51Ga), and (2) at $^{176}Hf/^{177}Hf$ = 0.280635 ± 0.000031 (n=14), corresponding to $\varepsilon Hf_{3.45\text{-}3.51Ga}$ = 4.2 ± 1.3, T_{DM}=3.51 ± 0.05Ga (8 zircons with ages between 3.45-3.51Ga).

6. Discussion

6.1 The Lower Onverwacht Group clastic meta-sediments

In this work, we report on the Hf isotopic composition of an uncommon occurrence of K-feldspar-rich meta-sediments from the Lower Onverwacht Group. This rock is petrologically similar to what Clemens et al. (2010) propose as the source of the Heerenveen batholith magmas, and we use it in the following discussion. However, the combined U-Pb-Hf isotopic compositions obtained from the detrital zircons of this rock deserve some comments in themselves.

Two populations of zircons are found in these arkoses, characterised by distinct $^{176}Hf/^{177}Hf$ values, plotting on two subparallel horizontal arrays between 3510 and 3300 Ma in the $^{176}Hf/^{177}Hf$ versus $^{206}Pb/^{207}Pb$ age diagram (Figure 3: supplementary online material). These arrays have two important implications, (1) they reveal that the detrital zircons were derived from two distinct sources within the crust and deposited from about 3510 Ma, and (2) that all ages <3510 Ma are likely, therefore to result from concordia-parallel Pb-loss (partial or complete). Pb-loss is commonly seen in Archean gneiss terranes, and is caused by multiple structural-metamorphic-magmatic overprints over a protracted period of time (for examples see Gerdes and Zeh, 2009; Zeh et al., 2009; Zeh et al., 2013;

Zeh et al., 2014). It is, therefore, probable that the clastic meta-sediments were deposited immediately after 3510 Ma, in general agreement with the interpretation of Dziggel et al. (2002), but that many grains underwent partial or complete Pb-loss during intrusion of the nearby Stolzburg and Theespruit plutons at ca. 3450 Ma and/or during subsequent structural metamorphic-magmatic events, which affected the BGS at c. 3.33 Ga, 3.25-3.20 Ga and c. 2.6-2.7 Ga (Details in Cutts et al., 2014; Heubeck et al., 2013; Schoene and Bowring, 2010). The grains with $^{207}Pb/^{206}Pb$ ages >3500 Ma always show lower U+Th contents (<250 ppm) compared to the younger grains (up to 600 ppm; see Data Item DR3). Thus it is possible that the younger grains accumulated more radiation damage compared to the older ones, and consequently were more metamict and sensitive to post-growth alteration. Nevertheless, a somewhat younger deposition age for the clastic meta-sediments, between 3510 and 3450 Ma, cannot completely be excluded. The only field constrain is that the Lower Onverwacht Group is intruded by the ca. 3450 Ma plutons of the southern BGGT (Anhaeusser et al., 1981; Moyen et al., 2018).

Questions remain regarding the source(s) of the two zircon populations in the Lower Onverwacht arkoses. As shown in Figure 9, the Hf isotope composition of both populations overlaps with data obtained from detrital zircons in Paleoarchean clastic sedimentary rocks (greywackes, quartzites and metapelites) of Swaziland in the Luboya-Kubuta river area (Taylor et al., 2016), and from the Dwalile Greenstone Belt (van Schijndel et al., 2017). This suggests that all these sediments originated from similar sources such as felsic rocks of juvenile character that occurred relatively widespread during the Paleoarchean. Nevertheless, their source(s) remain enigmatic. The hafnium-age signatures exclude coeval juvenile mafic rocks of the BGB, but also more felsic rocks of the Ancient Gneiss Complex, and the Lower Onverwacht volcanics. The mafic rocks, in particular the fine grained basalts as exposed in the Overwacht group, would supply no or few zircon grains, and most felsic rocks have lower εHf_t values (see Figure 9a). The very radiogenic composition ($\varepsilon Hf = +4$) of some of the zircon grains points to the existence of an early depleted mantle reservoir in the southeastern Kaapvaal craton, a feature also evident in mafic rocks from the area (Hoffmann and Wilson, 2017; Nebel et al., 2014; Schneider et al., 2019; Wilson et al., 2003). In addition, it requires that juvenile mafic rocks were transformed into felsic rocks immediately after their extraction from the mantle, either by fractional crystallisation within oceanic plateaus (similar to processes in the Rooiwater Complex of the Murchison Greenstone Belt further North: Laurent et al., 2019) or by melting of hydrated mafic crust.

6.2 The sources of the GMS suite

Compared to the TTG granitoids, the GMS suite has been less well studied. Early studies do not describe the range of rocks found in the batholiths, but are rather concerned with the dominant,

« core » phases of both Mpuluzi and Heerenveen batholiths, a justifiable choice given the relative volumes but nonetheless one that misses part of the story. Early work (Anhaeusser and Robb, 1983b; Robb et al., 1983), reiterated by Cornwright et al.(2008) and Robb et al. (2007), proposed that the GMS granitoids formed by melting of the TTG basement at crustal depths. This model is the underlying concept in more recent studies (Zeh et al., 2009) who described the post-3.2 Ga granitoids in Barberton area as resulting from « recycling » of older crust, but without detailing the petrogenetic process involved.

Clemens et al. (2010) offered a dissenting view. Based on experimental results (Watkins et al., 2007), they concluded that melting of TTGs cannot generate melts as potassic as the GMS suite, and that a more potassic source is required. They also noted the various rock types in the Heerenveen batholith and proposed that they reflect the implication of sources ranging from mafic to felsic, potassic igneous rocks or volcaniclastic rocks derived from such lavas (i.e. analog to modern subduction-related basalt-andesite-dacite-rhyolite series, similar to some components of the Fig Tree Group, and themselves formed by melting of a metasomatized mantle), all melting to produce the granitoids.

Here we revisit these findings in the light of our new isotopic constrains.

6.2.1 Different magmatic series

The GMS suite is characterized by a wide range of compositions, which correspond to different types of granitoids – possibly reflecting different magmas and sources.

Granitoids are not necessarily chilled melts but may represent the product of more complex magma chamber processes such as melt extraction from a solidifying mush (Bachmann and Bergantz, 2004).The texture of some of the GMS rocks, that of the syenites in particular (Figure 3d, I, j) certainly suggests that they might be the products of feldspar accumulation. Nonetheless, it seems difficult to use this as an explanation for the whole range of compositions featured in the GMS suite. In this case, one should observe an array trending towards the composition of K-feldspar, which is clearly not the case. The different trace element patterns between rock types also do not support such a model. It seems difficult, therefore, to propose that all the diverse GMS rock types reflect differentiation from a common parent, and we propose that the different rock types correspond to different magmas. Despite this feature, it is also apparent that the GMS suite exhibits homogeneous initial Hf isotope compositions, and petrogenetic models therefore need to accommodate a process that can explain this paradox.

6.2.2 The origin of the low-Ca granitoids

With weakly peraluminous composition, no clear differentiation trend (and an overall high-SiO_2 nature), as well as generally high levels in incompatible elements and negative εHf_t values, there is little that can relate the low-Ca granitoids to differentiation of mantle-derived mafic magmas (as proposed for instance by Kleinhanns et al., 2003). Rather, models involving melting of crustal lithologies from the BGGT are considered.

Figure 10 includes only samples from a geochemically homogeneous group (the low-Ca granitoids), thus reducing the impact of different source compositions; and uses tracers that are likely not controlled by accessory minerals. A first observation is the scatter of compositions in all the diagrams. This effectively rules out a single process forming a unique magmatic series. The origin of the low-Ca granites is thus composite.

From the laws of trace elements behaviour, the shapes of the trends generated by partial melting and fractional crystallization are expected to differ (Janoušek et al., 2016, chapter 21). In *log(compatible)* vs *log(incompatible)* diagrams (Figure 10a), partial melting generates shallow trends and fractional crystallization steeper ones (e.g. Robb, 1983). Both are evident for different sub-groups of the low-Ca granitoids. In *X/Y* vs. *X* diagrams, where *X* and *Y* are both incompatible (Figure 10b), fractional crystallization generates flat trends and melting forms sloped trends (Minster and Allègre, 1978). Again, both are evident in the low-Ca granitoids. In particular, the high-K_2O leucogranites and megacrystic granites from the Mpuluzi batholith depict rather clear fractionation trends whereas the low- and medium-K_2O granitoids seem to be governed by partial melting processes.

Th—U and K—Th diagrams (Figure 10c and d) do not allow such a clear discrimination of processes, but are nevertheless good tracers of the sources or the initial magmas (Cuney et al., 1989; Friedrich et al., 1987). Here it is clear that the low-Ca granitoids do not generate a single trend, but rather require parallel evolution from a range of primitive magmas, themselves originating from diverse sources.

Collectively, the low-Ca granitoids therefore appear to result from (i) melting of compositionally varied (although broadly similar, as they have similar major element properties) sources and (ii) fractionation of some of the magma batches. This is, of course, a rather common scenario for the formation of crustally-derived granites/leucogranites (Clemens et al., 2009; Cuney et al., 1989; Stussi, 1989; Turpin et al., 1990). The feasibility of various lithologies in the BGGT as sources for these rocks is now considered:

Mafic rocks. From a purely isotopic point of view (Figure 9a), mafic rocks may be possible sources for the GMS granitoids. However, ample petrological evidence in particular from experimental petrology (reviews in e.g. Hoffmann et al., 2018; Moyen and Stevens, 2006) demonstrates that melting of mafic rocks does yield potassic liquids but rather sodic (TTG) magmas. TTGs are indeed found in the BGGT and in Swaziland at 3.33 – 3.22 Ga and their isotopic characteristics perfectly fit the crust evolutionary trend defined by the $^{176}Lu/^{177}Hf$ ratios of the mafic rocks (Figure 9a). Thus, we rule out common mafic rocks of the BGB as a possible source for the low-Ca granitoids.

Intermediate supracrustal rocks. Clemens et al. (2010) propose that the dominant rock types of the Heerenveen batholith form by melting of volcanic or volcaniclastic rocks rather than the commonly assumed TTGs. Such rocks are more potassic and more hydrous than typical TTGs, and are thus a better source to form large volumes of potassic magmas. Several sedimentary or volcano-sedimentary units are known in and around the BGB, although generally in small volumes: (i) components of the ca. 3530 Ma Lower Onverwacht Group; (ii) large parts of the ca. 3250 – 3300 Ma Fig Tree Group; (iii) the ca. 3210 Ma Moodies Group.

The Lower Onverwacht Group occurs as fragments in the amphibolite-facies terrane south of the BGB (the Stolzburg block), where it occupies a very minor area. It is dominated by amphibolites with subordinate felsic schists, interpreted as former volcanic and volcaniclastic rocks, with very minor clastic metasediments. The protoliths of the felsic schists of the Theespruit Formation erupted ca. 3530 Ma ago, and have εHf_t between -3 and +1. Evolution from these values, with the appropriate bulk rock composition ($^{176}Lu/^{177}Hf$ = 0.005 to 0.009) projects to εHf_t of -4 to -10 at 3100 Ma, below the compositions of the GMS suite. Zircon from the Theespruit clastic meta-sediments reveal two clusters, both with suprachondritic εHf_t between +2 and +4 at ca. 3510 Ma. Using an average $^{176}Lu/^{177}Hf$ ratio for continental rocks of 0.0113, they project to -2 to -4 at 3100 Ma, i.e. on the lower end of the GMS granitoids.

A higher $^{176}Lu/^{177}Hf$ ratio would allow an evolution to εHf values of ca. -1 at 3100 Ma, overlapping with the GMS magmas. This requires $^{176}Lu/^{177}Hf$ ratios higher than those observed in Barberton felsic rocks (0.015), which is possible assuming that some of the detrital zircon grains survived partial melting, consequently increasing the apparent $^{176}Lu/^{177}Hf$ ratio of the extracted melt. However, this seems at odds with the lack of inherited grains with ages >3200 Ma in the GMS rocks, and with relatively elevated Zr contents (and thus zircon saturation temperatures in excess of 800 °C). At more than 800 °C, clastic meta-sediments would melt 30-40 % (Vielzeuf and Montel, 1994), rendering zircon preservation unlikely. Thus, arkoses of the Lower Onverwacht Group may represent a source for the GMS magmas but only if one assumes the superposition of a variety of favourable

circumstances. Such arkoses are also limited in extent making it difficult to account for the widespread melting that must have taken place to accommodate the high volumes of melt required for GMS emplacement.

The Fig Tree Group contains felsic volcanic and volcaniclastic rocks as well as immature sandstones (Toulkeridis et al., 1999). Available Hf isotope data shows that zircons from the Fig Tree Group plot in a wide range, bound by the clastic meta-sediment evolution line. Felsic volcanic and volcaniclastic rocks represent a petrologically favourable lithology, very appropriate to generate GMS magmas (Clemens et al. 2010), and they are more abundant volumetrically than the Lower Onverwacht arkoses, but they still represent low volumes regionally. Furthermore only the combination of the most favourable circumstances (both $^{176}Lu/^{177}Hf$ ratio and original εHf_t values at the outer end of the observed range) could produce GMS-like compositions.

The same isotopic reasoning applies to the ca. 3210 Ma Moodies Group. Although the Moodies Group forms large volumes of rocks, only a small portion is made up of greywackes, volcaniclastic or volcanic rocks: the bulk of the Group is made of non-fertile quartz-rich sandstones.

In summary, from (isotope) geochemical points of view it is permissible but unlikely to generate the GMS magmas by melting of regional metasediments or metavolcanics. Furthermore, no know geological unit includes sufficient volumes of such rock types to generate the large volume of GMS granitoids that are preserved.

Meta-plutonic rocks. The conventional view on the formation of Archaean potassic granites is by melting of the volumetrically dominant TTG granitoids or meta-TTG gneisses that dominated the crust pre-3.1 Ga. The composition of the TTG basement around the BGB and Swaziland is well established. Using reasonable $^{176}Lu/^{177}Hf$ values, the older (ca. 3.53 Ga and ca. 3.45 Ga) generations of TTGs are not radiogenic enough to represent a potential source for the GMS magmas. On the other hand, the 3.28 – 3.23 Ga TTGs area better match. They span a very large range of εHf values (-2 to +3), that projects to about -4 to +1 at 3100 Ma, overlapping the range of GMS magmas. This source is also supported by the finding of zircon xenocrysts with ages of about 3200 Ma in some GMS samples. From a purely zircon isotopic perspective, they represent, therefore, the most likely source.

To some degree, this is in contradiction with the findings of Clemens et al. (2010), who found that TTGs are not an appropriate source for the GMS magmas. However, their argument is chiefly based on the low abundance of biotite in ordinary tonalites to trondhjemites of the TTG suite compared to more fertile rocks, therefore preventing the formation of large volumes of magmas at reasonable temperatures, and the formation of sufficiently K-rich melts (Watkins et al., 2007). However,

amongst the ca. 3.2 Ga TTGs, large volumes (in particular the Kaap Valley Pluton, the largest around the BGB; and portions of the Nelshoogte Pluton) are made of a relatively mafic (SiO_2 ≈ 60-65 %) tonalite to leucodiorite (Moyen et al., 2018), a rock that is mafic enough to contain large volumes of hydrous silicates such as biotite and amphibole. In fact, a mafic tonalite such as the Kaap Valley has a composition very close to a greywacke or an andesite, and thus would work just as well as a petrological source for the GMS rocks. Tonalites are also readily available in the BGGT, where they form large volumes of exposed rocks in the NW Barberton terrane, and occur more sporadically e.g. in Swaziland (part of the Usutu suite).

Combination of the above. An obvious problem with the GMS suite is the volume of source required. Assuming a conservative 20% melting of the source to form granitoids, a volume of source 5 times bigger than the granitoids is required. The present day Lower Onverwacht and Fig Tree Groups represent only small relicts at the surface. It is unlikely that they once formed effectively the bulk of the present day 100 x 200 km domain where the GMS granites occur. In fact, proposing any lithology to be homogeneous on that scale is not very realistic, which is in good agreement from our observations on the geochemistry of the low-Ca granitoids that require multiple continental sources (Figure 10).

The middle and lower Archaean continental crust is made up of Grey Gneisses, a complex lithological assemblage made of many tectonically transposed and interleaved types of rocks, dominated (but not restricted to) TTGs. Grey Gneisses however also include other components. To take one example, the nearby Ancient Gneiss Complex of Swaziland includes components that cover a range of composition much wider than simple TTGs (Hofmann et al., 2019; Hunter et al., 1978). Mid-crustal composite Grey Gneiss therefore forms a suitable source for the low-Ca portion of the GMS batholiths. It supplies a range of isotopically related (or similar) rocks with varying chemical compositions. Depending on the exact proportions of various components in the source rock, the resulting magmas could cover a range of compositions, all more potassic than the protolith. The sediments interleaved in the orthogneisses would of course derive from the TTG component in which they now occur and thus have the same isotopic (and trace elements, largely) properties, and amphibolite enclaves could be a further source of water stored in amphibole or biotite.

6.2.3 The origin of the syenites and related rocks

The origin of syenites is, generally, a controversial topic. A first issue is that many could be K-feldspar cumulates rather than chilled liquids, and indeed the textures of GMS syenites (Figure 3i, j) is consistent with this hypothesis. Even so, the formation of K-feldspar, quartz-free cumulates requires a liquid of an unusual, potassic and low silica, composition.

Like other syenites in the world (Harris et al., 1983; Tchameni et al., 2001; Woolley and Jones, 1987; Zanvilevich et al., 1995), the parental magma of the GMS syenites has been related to deep crustal melting (Anhaeusser and Robb, 1983b). However, in order to generate a low-silica, high-potassium liquid (rather than an A-type granite: Creaser et al., 1991; Skjerlie and Johnston, 1992), an already mafic and potassic crustal source is required, for instance a K-rich arc-like basalt (Clemens et al., 2010). Compared to experimental compositions, the syenitic component indeed plots in the field of melts formed by melting of high-K mafic compositions (Figure 11a; Laurent et al., 2014a). An issue with this model is the complete lack of such a component in the stratigraphy of the Barberton Greenstone Belt. However, from a geochemical point of view, most of the supporting evidence can also be interpreted in favour of differentiation of a similar K-rich basalt. This model is similar to what is generally offered for Phanerozoic high-K calc-alkaline granitoids (Couzinié et al., 2016; Fowler et al., 2008; Fowler and Rollinson, 2012) and Archaean sanukitoids (Laurent et al., 2014a; Laurent et al., 2014b; Martin et al., 2010). In these examples, the parental mafic component occurred as small volumes only and is seldom preserved in itself, but is invariably hybridized and differentiated, and is preserved only as small enclaves, dykes, etc. in the granitoids. Similar hybridization processes may account for the absence of a pristine high-K mafic component in the BGGT.

The syenitic component of the GMS suite offers similarities with the late-Archaean sanukitoids, in particular its K-rich nature for low silica contents, and the high Sr+Ba amounts. This is well expressed in the “FSMB” (= [FeOt + MgO] * [Sr + Ba]) component in the diagram of Laurent et al. (2014a) (Figure 11b). Sanukitoids are commonly associated temporally and spatially with syenites (Bibikova et al., 2006; Fayol and Jébrak, 2017; Heilimo et al., 2012; Lobach-Zhuchenko et al., 2008; Sutcliffe et al., 1993).

Sanukitoids (and related rocks) are potassic as a result of enrichment of the mantle source by small volumes of crustal components such as sediments, during lithosphere recycling previous to the melting. Given the imbalance of Hf (and also Sr or Nd) concentrations between sediments and the mantle, a small amount of contaminant (5-10 %) will effectively fully control the isotopic composition of the hybrid source (Couzinié et al., 2016). Since the sediments ultimately are derived from the local crust, the isotope composition of the resulting magmas is effectively undistinguishable from that of pure crustal reworking (Laurent and Zeh, 2015).

In the case of the GMS suite, the syenitic component would thus be related to burial of small portions of the local crust (with the characteristics we described previously) into the mantle, presumably during the ca. 3.2 Ga “orogenic” event (Moyen et al., 2006; Moyen et al., 2018; Schoene and Bowring, 2010). This would form a suitable mantle source for high K mafic magmas at ca. 3.1 Ga.

These magmas would subsequently inject the partially molten Grey Gneiss crust, differentiate and interact there, locally forming K-feldspar-rich cumulates representing the present-day syenites.

6.2.4 The origin of the dark granitoids

The dark granitoids are less common and less well constrained than the other two groups. They are relatively low silica, and are diverse in composition. Tentatively, we propose that they are also related to melts from the enriched mantle (much in the same way as the mafic components of the syenite group). In the case of crust-mantle hybrids such as envisioned here, the main control on the trace element composition of the liquid is the *F/a* ratio, namely the balance between the degree of melting (*F*) and the proportion of contaminant (*a*) (Moyen, 2009; Moyen, 2019). In this view, the smaller degree of incompatible element enrichment of the dark granitoids could be ascribed to a higher *F/a* ratio corresponding either to a lesser amount of mantle contamination or to a higher degree of melting. Since the crustal contaminant was the same material, the hybrid mantle source also inherited crustal characteristics.

The diversity within the dark granitoids also reflects partial melting of a heterogeneous mantle, with more or less enriched portions, related to input by crustal material with varying composition (sediments, igneous felsic rocks, etc.) during >3.1 Ga orogenic events. .

6.3 On the isotopic homogeneity of the GMS granitoids

The large size of the GMS batholiths, in addition to the petrological evidence presented above, strongly suggests a composite source for these rocks with different rock types melting. The low-Ca granitoids, that form the bulk of the Suite, are interpreted as melting of (felsic) crustal lithologies. In contrast, the dark granitoids and, probably, the syenites formed by melting of mantle (metasomatized by crustal additions). The isotopic evidence is apparently at odds with this conclusion. Thus, a process generating a range of petrologically distinct, but isotopically identical sources is required. The problem is actually slightly different at the crust, or lithosphere scale (Figure 12).

At the scale of the lithosphere, we have a range of magmas, from different sources (crust or mantle) that nevertheless have a similar isotopic signature. A similar situation has been described in several post-orogenic settings (Couzinié et al., 2016; Laurent and Zeh, 2015; Payne et al., 2016). The key point of these settings is that the mantle that melts and contributes to the granitic magmatism has been enriched by material buried from the local crust. Thus, the Hf in both the crust and mantle-derived magmas ultimately has the same source and the same characteristics.

At the scale of the crust, the problem is less severe. In fact, it is geologically unrealistic to imagine that rocks with the exact same isotopic features existed over several tens or hundreds of square kilometres. Thus, some process of intracrustal isotopic homogenization must take place. This interpretation is supported by the facts that all GMS granites (a) have the same isotopic composition; (b) were emplaced over the same, relatively short time interval, and (c) fit on the same crust evolutionary trend as TTGs and granites of the BGGT and Swaziland younger than 3400 Ma, as well as sediments from the Fig-Tree and Moodies Groups (but not pre-3400 Ma rocks)(Figure 9). The last point suggests that Hf isotope homogenization occurred within the deep to middle crust either prior to, or during GMS granitoid formation at ca. 3100 Ma. Lower- to mid-crust homogenization is further supported by similar Hf isotopic compositions of granitoids with ages of ca. 2700 Ma, which fall on the same crustal evolutionary trend as the GMS granites (Figure 9a). Hafnium isotope homogenization perhaps was achieved by percolation of melts and hydrous fluids in the deep hot crust (Kröner et al., 2018). Fluid and melt circulation allows the dissolution of the original zircon populations (magmatic or detrital), followed by Zr-Hf-transport, and Hf isotope homogenization, prior to new zircon precipitation (Zeh et al., 2015; Zeh et al., 2020). Evidence for Hf isotope homogenization during amphibolite-facies metamorphism in the middle/lower crust of the AGC is reflected by the metamorphic zircon grains found in rocks of the Dwalile GSB (van Schijndel et al., 2017).

The homogenization process in the middle crust provides insight into a poorly understood aspect of Archaean geology, namely the incorporation of igneous rocks of the TTG suite into complex Grey Gneisses.

Here, we emphasize the difference between “TTGs” and “Grey Gneisses”. TTGs are igneous plutonic rocks. Grey Gneisses, on the other hand, are complex units that have long been identified as forming the bulk of the Archaean crust (Hunter, 1970; Salop and Scheinmann, 1969). Grey Gneisses commonly include (meta-)TTG components, possibly formed during several plutonic phases – in some cases hundreds of million years apart (Kröner et al., 1999; Laurent et al., 2019). They also include a range of other lithologies such as amphibolites or occasionally metasediments. All these rocks have been tectonically interleaved, deformed, transposed, metamorphosed and often partially molten in the middle or lower crust (Kröner et al., 2019; Kröner et al., 2014; Kröner et al., 2018; Laurent et al., 2019; Taylor et al., 2013; Taylor et al., 2020). In the Kaapvaal Craton, Grey Gneisses correspond e.g. to the Ancient Gneiss Complex of Swaziland (Hunter, 1970; Kröner et al., 2019), or the dominant rock type in the Northern Kaapvaal (Laurent et al., 2019). The processes by which TTG plutons are incorporated in Grey Gneiss complexes are not elucidated, but at least require deformation and burial.

Returning to the GMS suite, it is proposed that these granitoids are the product of melting of Grey Gneisses – but not of TTGs.

6.4 Crustal stabilization in the Archaean

The GMS granitoids were emplaced at a key period in the evolution of the central portion of the Kaapvaal craton (i.e. south the the Thabazimbi-Murchsion Lineament, TML: de Wit et al., 1992). They immediately follow (and conclude) a period of "permobile" regime, with the formation of classical granite-greenstone terrain, important (TTG) magmatism, metamorphism and deformation of the crust. Ca. 3.1 Ga ages actually occur throughout the craton (e.g. Poujol et al., 2003), evidencing a craton-wide process.

After the emplacement of the GMS suite, however, the geological activity in the Kaapvaal craton in general and in Barberton in particular changes radically. The first, large intracratonic sediment accumulations (Pongola supergroup at ca. 2.95 Ga; Wilson and Zeh, 2018) cover the craton shortly after this episode, and further geological evolution in the Central Kaapvaal craton has a different character – localized, brittle intrusions of mafic rocks (Klausen et al., 2010) and "late" granitoids (Meyer et al., 1994; Robb et al., 2006). The now rigid proto-Kaapvaal Craton acts as a rigid nucleus against which late-Archaean mobile belts in the North (Laurent et al., 2019; Vézinet et al., 2018) and West (Schmitz et al., 2004) are accreted.

The geochemistry of the GMS suite shows that the process of craton stabilization involved the lithosphere as whole (mantle as well as crust), and that this lithosphere had to be primed by a pre-existing history, evident in the study area by events at ca. 3.2 Ga (Schoene et al., 2008; Schoene et al., 2009). The purpose of this paper is not to comment on the controversy relating to the ca. 3.2 Ga geodynamics of the BGGT. From a purely petrological point of view the subsequent formation of the ca. 3.1 Ga GMS suite requires (i) the formation of an enriched lithospheric mantle, and (ii) the formation of composite Grey Gneisses in which diverse components are juxtaposed and partially equilibrated and homogenized. The melting of this lithosphere permits the formation of the GMS suite as a whole. In a sense, this is reminiscent of post-orogenic granite-forming environments (Bonin, 2004; Moyen et al., 2017), although this should not necessarily be taken as evidence for a modern-looking plate tectonic environment (Moyen and Laurent, 2018), as the requirements are merely the existence of a composite crust, and burial of crustal material in the mantle.

A key difference with modern settings is the timespan between the ca 3.2 Ga events and the formation of the GMS suite at 3.11 Ga. Whereas in modern orogenic belts collision and post-collision are separated by a few millions to tens of million years, with the two stages blurring into each other, here the two events are 100 Ma apart and very few ages are recorded between the two peaks: some

minor felsic dykes as well as the Dalmein pluton (Kamo and Davis, 1994; Lana et al., 2010; Zeh et al., 2009). In the GMS suite, rare zircon xenocrysts also record magmatic activity pre-3110 Ma. Unpublished data by Murphy (2015)(Figure 13) also suggest that a moderate igneous activity may have taken place from ca. 3.2 to ca. 3.1 Ga (Figure 13). Finally, there are some hints (Lana et al., 2011) that the Pigg's Peak Batholith in Swaziland also accreted over a long period of time between 3.2 and 3.1 Ga. If confirmed, this information would put the GMS suite in a new light, as the culmination of a long-lived history, itself the aftermath of the major ca. 3.2 Ga crustal accretion event. Thus, it appears that the BGGT crust was primed by the ca. 3.2 Ga evolution, then slowly matured until ca. 3.1 Ga, at which point massive amounts of melt were generated in, or extracted from the mid- to lower crust, likely triggered by processes in the mantle. This succession of events resulted in the formation of a very stiff, stable lithosphere that became the core of the Kaapvaal craton proper.

We further speculate that this process is a common occurrence in Archaean cratons. In several Grey Gneiss units, long-lived magmatic activity has been reported (Laurent et al., 2019; Taylor et al., 2020). This continuous or near continuous activity in deep crustal levels seems to contrast with the sharp peaks observed in granitoids emplaced in the upper crust. However, the latter age distribution corresponds to crystallization of granitic magmas in large batholiths, whereas the former reflects formation of small melt volumes. Integrated over the whole crust, the process may then be one of slow formation and accumulation of melt in the source regions, followed by brutal extraction and quick emplacement at upper crustal levels (Bons et al., 2004; Vanderhaeghe and Teyssier, 2001).

7. Conclusion

This study investigates the way the Archean lithosphere stabilizes and forms cratons, and sheds light on the evolution from early TTG-greenstone crust to more stable cratonic crust. The Archean protocrust consists of lavas and minor sediments in greenstone belts, intruded by TTG plutons. Whereas the petrological processes to form it have been studied and are reasonably well understood, this protocrust is not strong enough to permit the deposition of large clastic units and the injection of dense dyke swarms (Cawood et al., 2018).

Here, we uncover some of the processes that mature the protocrust into cratonic crust. The formation of a stable continental mass requires firstly the formation of meso-crustal Grey Gneiss complexes. Grey Gneisses are composite units. They include a significant portion of meta-TTG, but they also include meta-sediments, mafic rocks, melts, etc. Furthermore, they underwent a degree of homogenization through both mechanical (tectonic interlayering) and chemical (fluid percolation,

dissolution/crystallisation, diffusion) processes. Another necessary ingredient appears to be the burial of some of the crustal material into the mantle, to form an enriched mantle source (although one sharing its isotopic characteristics with the overlying crust). In the BGGT, this is the result of the ca. 3.2 Ga geological processes.

Subsequently, all portions of this lithosphere or proto-lithosphere melt, resulting in the formation of large, composite plutonic units with a diversity of compositions, in a situation reminiscent to what is observed in present-day post-orogenic domains. Crucially, this also permits scavenging of incompatible heat producing elements, which are carried closer to the surface, leaving behind a refractory, cold residue that has little potential for future reworking and forms the lower crustal nucleus of the stable cratonic lithosphere.

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This publication has been long overdue, as it is based on data acquired many years ago. It would have been an honour and a pleasure to discuss our findings with Alfred Kröner, who in addition of being a great geochronologist, was also extremely knowledgeable on the geology of the BGGT – a place very close to his heart, and where he conducted field work until his last days. Unfortunately we left the opportunity slip, and the best we can do now is to offer our contribution to this Special Issue commemorating his achievements. We thank L. Robb and J. Payne for helpful and constructive reviews.

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Data Availability

Datasets related to this article can be found at http://dx.doi.org/this/article, an open-source online data repository hosted at Mendeley Data (Moyen et al, 202X).

Item DR1: Whole rock analyses of granitoids from the GMS suite, compiled from this work as well as from the literature.

Item DR2: U-Pb analyses in zircon grains from the GMS suite and the Theespruit formation.

Item DR3: Lu-Hf analyses in zircon grains from the GMS suite and the Theespruit formation.

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Figure 1. Schematic map of the BGGT, highlighting the GMS batholiths. GMS syenites: Bk: Boesmanskop; KZD: Kees Zyn Doorns. TTG units: Bpl.: Badplaas Complex; Stz.: Stolzburg Pluton.

Figure 2. Geological map of the Heerenveen batholith and adjacent portions of the Mpuluzi batholith. See Figure 1 for location. After Moyen et al. (in press). The location of the samples out of which zircons were analysed in this study is reported with coloured symbols (colour code as in Figure 4). Samples from the literature are in white symbols. W05 : (Westraat et al., 2004), samples « phon » (Boesmanskop phonolite dyke) and 586 (Mpuluzi granite). K94 : (Kamo and Davis, 1994), sample S (Mpuluzi granite) and T (Boesmanskop syenite).

Figure 3. Field photographs of the main rock types in the Southern GMS batholiths. Coloured symbols next to the photos correspond to the caption of Fig. 4. (a—d) : Heerenveen : (a). Homogeneous megacrystic granite of the core portion of the batholith, here with sub-euhedral centimeter sized grains of quartz. (b) Leucogranite II, associated with pegmatites, in the South-Eastern Shear Zone (scale bar is about 10 cm). (c). Lit par lit injection of leucogranite I into gneissic remnants (near sample HE-11-4A) in Western shear zone. (d). Pink quartz monzonite (near sample HE-11-8). (e—i) : Mpuluzi : (e). Main homogeneous megacrystic granite of the Heerenveen batholith, here with a small microgranular mafic enclave. (f). Leucogranite II (scale bar is about 1 cm). (g). Intrusive breccia of a dark Augen granodiorite (Aug) brecciating a syenogranite (SG) with early pegmatites (Peg). Scale bar is about 1 m. (h). Augen granodiorite. (i). Syenogranite. Photos f—i come from a large outcrop (the Eagle High locality) mapped in details by Sonke (2006), C. Campbell and A. Kisters (unpub), further described in Moyen et al. in press, and from which samples MP-11-2 (syenogranite), MP-11-3 (Augen granodiorite) and MP-12-01 (pegmatite) were taken. (j—k) : Boesmanskop : (j). Coarse grained syenite. (k). Hypovolcanic dyke (equivalent to sample phon of Westraat et al. 2004).

Figure 4. Major-elements geochemical features of the GMS batholiths of the Southern BGGT. Colours reflect the different chemical types (and field-based facies), and symbols the different intrusions. Samples studied for U—Pb—Hf in zircon are shown as larger symbols. (a) P—Q (cationic) diagram of (Debon and Lefort, 1983). (b) SiO_2 – MALI (weight) diagram of (Frost et al., 2001). (c) A/CNK vs. A/NK diagram of (Shand, 1943). A/CNK = molecular Al/ 2Ca + Na + K and A/NK = molecular Al/Na+K (or equivalent oxide-based formulations). (d) B—A (cationic) diagram of Debon and Lefort (1983). (e) B – mg# diagram of Debon an Lefort (1983), mg# = (cationic) Mg/(Mg+Fe). (f). SiO_2 – K_2O (weight) diagram, with lines corresponding to the series defined by Peccerillo and Taylor (1976).

Figure 5. TTG normalized spidergrams (comments in text). Tm is omitted, as it is not reliably analysed in our dataset, and Lu and Yb are swapped to obtain smoother patters; this reflects the hook-shaped REE pattern of TTGs, with LuN > YbN. Colours as in Figure 4, samples dated in this work appear as thicker lines. The grey fields show the whole range of our data, for orientation.

Figure 6. TTG normalized spidergrams for various trace elements, arranged by increasing variablity. Comments in text, same caption as Figure 5.

Figure 7. Zoning patterns of zircons from the Heerenveen and Mpuluzi batholiths, and from Lower Onverwacht volcanics and meta arkose. Circles mark laser spots drilled for U-Pb dating; sample number and respective $^{207}Pb/^{206}Pb$ ages are presented as well. Most images are back-scattered electron images, cathodoluminescence images are marked with CL.

Figure 8. Representative concordia diagrams showing results of U-Pb dating of magmatic zircon in different samples of the (a-c) Heerenveen batholith, (f) Mpuluzi batholith, and in (e) Theespruit meta-volcanics, as well as of (f) detrital zircons in Lower Onverwacht meta-arkose. (Concordia diagrams of all investigated samples are shown in Figure 2 : supplementary online material).

Figure 9. Age-εHf diagram summarizing the results of U-Pb-Hf isotope analyses of (a) felsic and (ultra)mafic igneous rocks, and (b) clastic metasedimentary rocks from the Barberton Greenstone Belt (GSB), Swaziland, and Commondale GSB. Data source: (1) Zeh et al. (2009), (2) Zeh et al. (2011), (3) Zeh et al. (2013), (4) Kröner et al. (2013), (5) Kröner et al. (2014) (6) Kröner et al. (2016) (7) Kröner et al. (2018), (8) Hoffmann et al. (2017), (9) Hoffmann et al. (2016) (10) Taylor et al. (2016), (11) van Schijndel et al. (2017), (12) Schneider et al. (2019). M-Mpageni granite, SMN- Sincunusa - Mbabane-Ngwempisi-granites, S-Sinceni granite, C-Cunningmore tonalite, MNP-Mpuluzi-Nelspruit-Piggs Peak batholithe (i.e. GMS suite), KNS-Kaap Valley-Nelshoogte-Stentor-tonalites, D-Dalmein granite, AGC-Ancient Gneiss Complex-granite dikes. CHUR-chondric uniform reservoir, DM-depleted mantle evolution based on $^{176}Hf/^{177}Hf$(today)=0.283250, and $^{176}Lu/^{177}Hf$=0.0384 (Chauvel and Blichert-Toft, 2001); Arrows mark crust evolutionary trends [$^{176}Lu/^{177}Hf$=0.0113, average continental crust; average (Taylor and McLennan, 1985) and (Wedepohl, 1995); $^{176}Lu/^{177}Hf$=0.026; average of Barberton ultramafic rocks, n=48; from Schneider et al. (2019).

Figure 10. Trace element characteristics of low-Ca granitoids of the GMS suite. Symbols and colours as in Fig. 4. On each diagram, the overall evolution is related to partial melting (PM) and fractional crystallization (FC), as discussed in the text and indicated by grey arrows.

Figure 11. Discussion on the syenitic component is based on the two diagrams of Laurent et al. (2014a). (a) A ternary diagram Al_2O_3/(FeOt+MgO) – 3 CaO – 5 K_2O/Na_2O offers a good discrimination between melts generated from different (crustal) sources.. (b) the A/CNK (= molecular Al_2O_3 / CaO + Na_2O + K_2O) – Na_2O + K_2O – 2 FSMB (= [FeOt + MgO] × [Sr + Ba]) ternary diagram efficiently isolates the "sanukitoid-like" component, pointing towards the FSMB apex, from other types of granitoids.

Figure 12. Summary of the petrogenetic processes leading to petrological diversity but isotopic homogeneity.

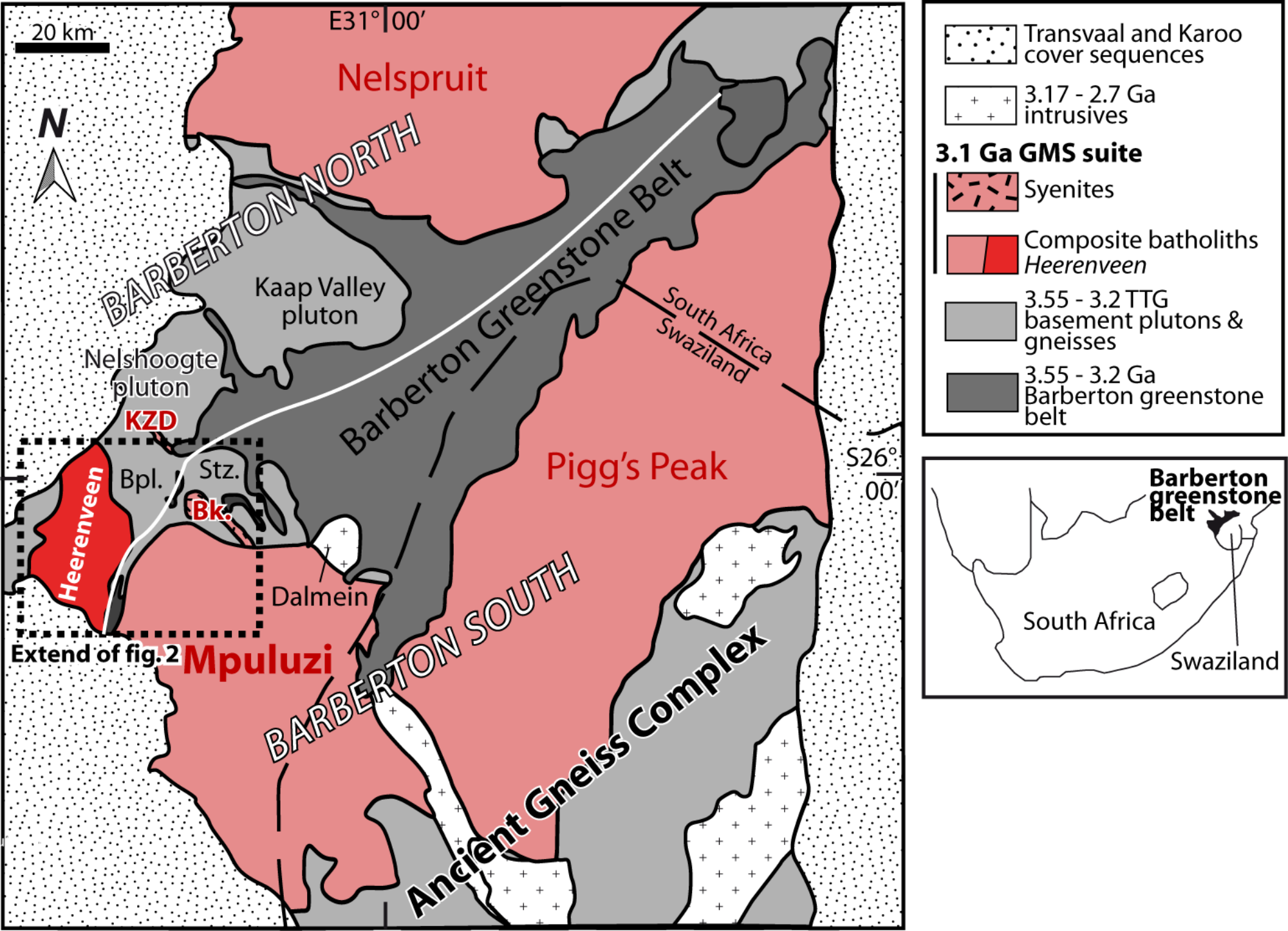
Figure 13. Age vs K_2O. In addition to ages reported in Table 3, this figure also includes (unpublished) ages by Murphy (2015), depicted as smaller symbols (with the same colour code). Symbols as in Figure 4. This evidences the possible presence of long-lived pre-3110 Ma magmatism (high K_2O, low Ca granitoids) that we attribute to low degrees of melting (± fractionation) of the Grey Gneisses, contrasting with the short pulse of very diverse magmas of the GMS event proper at ca. 3.1 Ga, witnessing higher degree of melting of a range of sources. Inset: distribution of published ages in the BGGT (top) and AGC (bottom). Both feature the same, ca. 3230 Ma peak. However, in the deeper structural levels (the ACG) , note the occurrence of 3150—3200 Ma ages that attest to the persistence of hot (melting?) conditions in the lower/middle crust during this period. The ca. 3.1 Ga peak in the upper crust (BGGT) reflects the extraction and emplacement of all melts in the GMS suite.

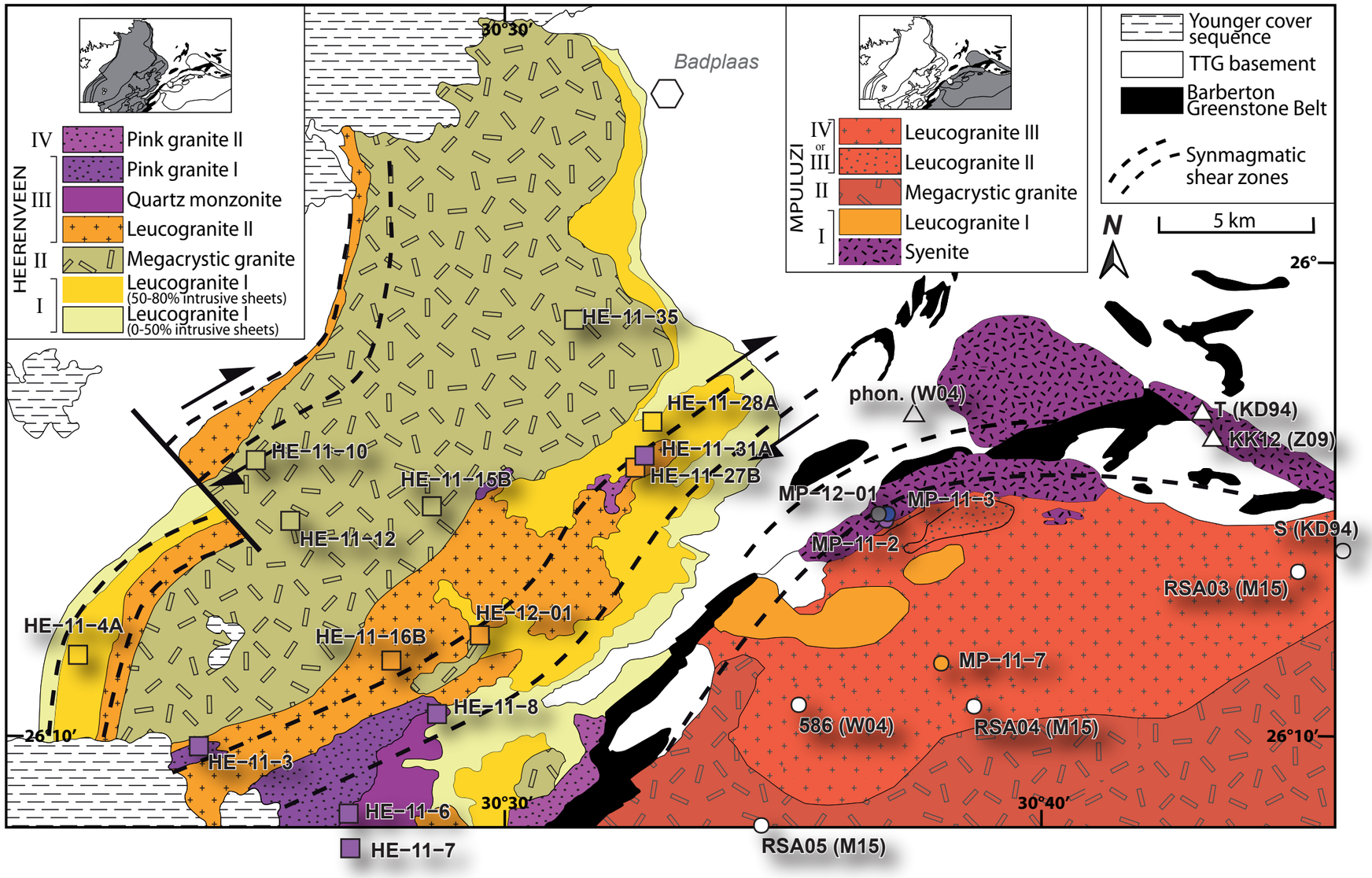
Table 1. Summary of the chemical types of GMS rocks. The types and sub-types (first two columns) are those defined in this work, see text and Figure 4. The matching facies in each batholith are indicated (Figure 2). The chemical description from the last columns is given on the basis of the key features identified by Bonin et al. (2020). calc-alk: calc-alkalic; alk-calc: alkali-calcic; alk: alkali, terminology after Frost et al. (2001).

Table 2. Normalizing factors and order used in the diagrams of Figures 5 and 6.

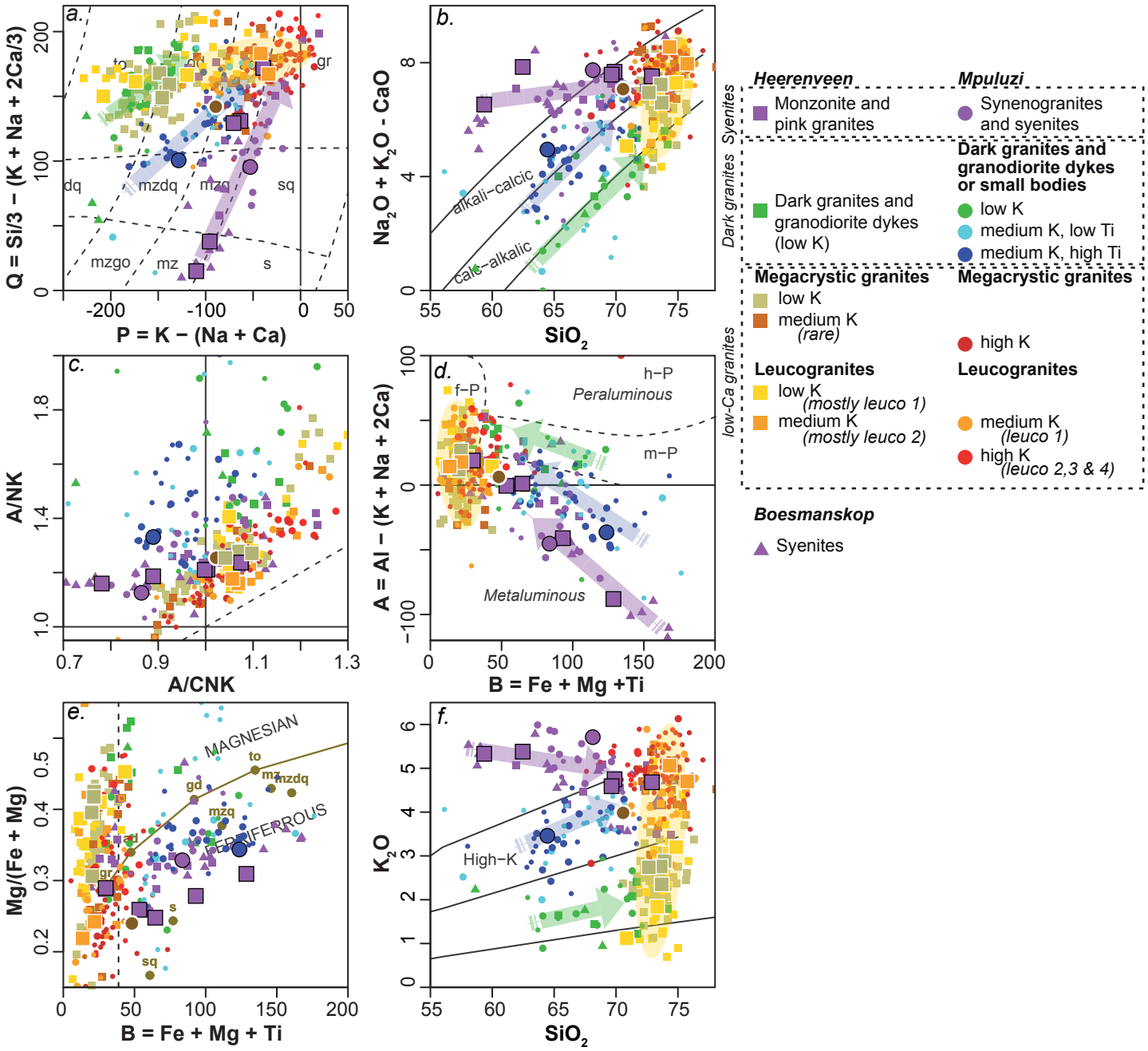
Table 3. Summary of U-Pb ages obtained in this study on the Heerenveen and Mpuluzi batholiths, and on meta-volcanic rocks of the Lower Onverwacht Group, compared with published data (shaded rows). Long. and Lat.: longitude and latitude, given as decimal degrees.

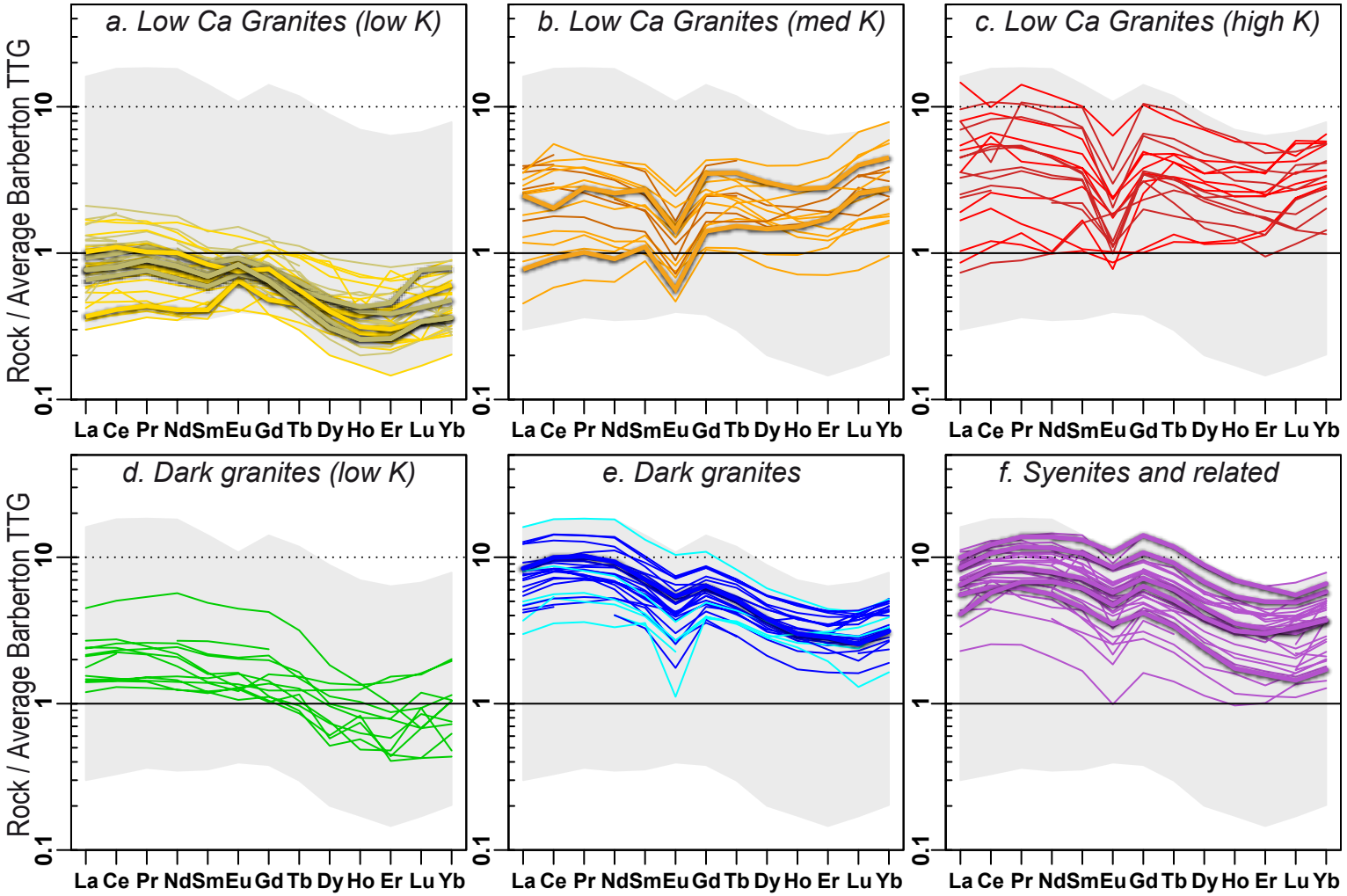
Table 4. Summary of Hf isotopic results obtained in this study.

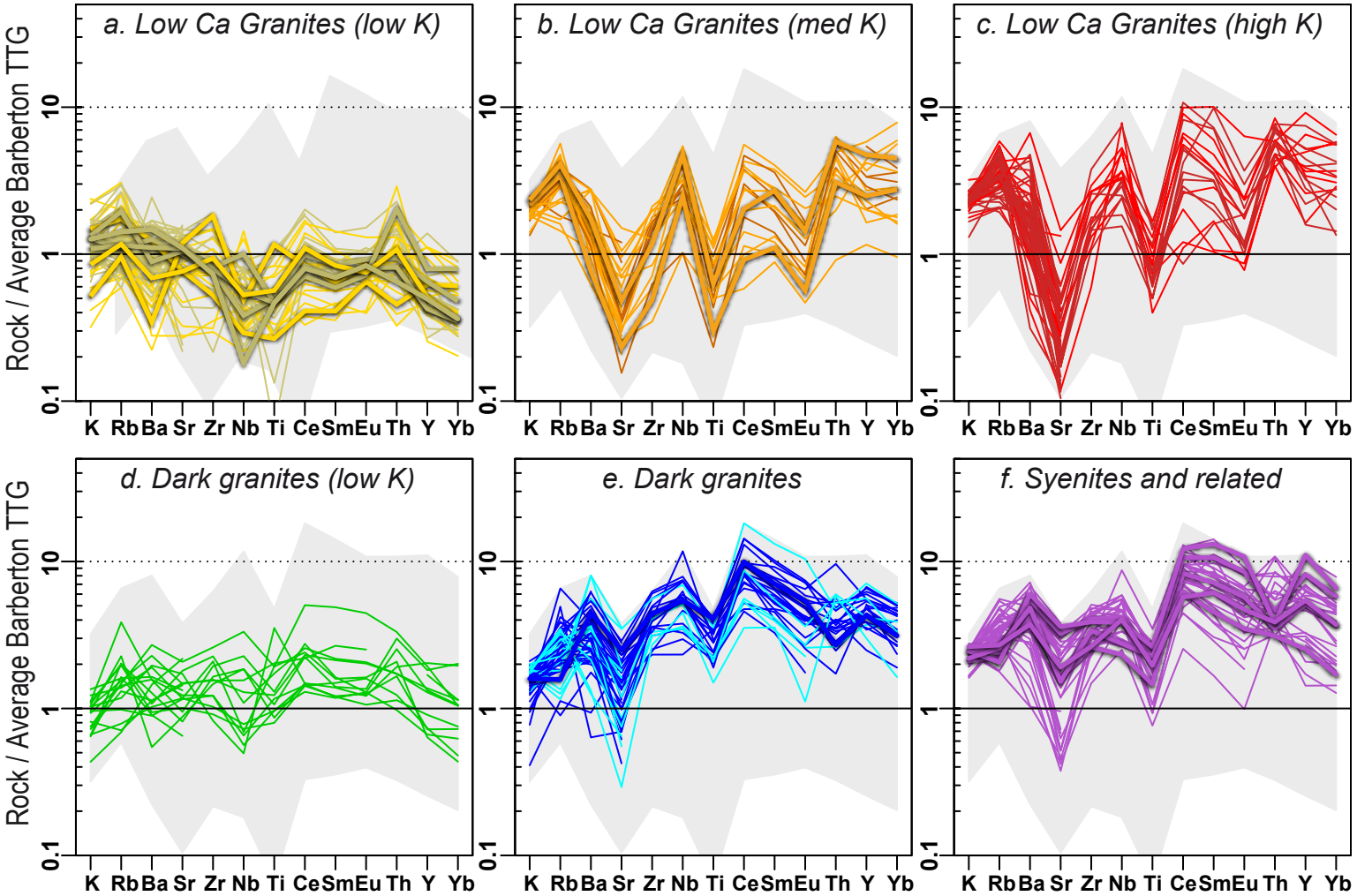

20 km
N
E31°00'
Nelspruit
BARBERTON NORTH
Barberton Greenstone Belt
Kaap Valley pluton
Nelshoogte pluton
KZD
Heerenveen
Bpl.
Stz.
Bk.
Dalmein
Extend of fig. 2
Mpuluzi
BARBERTON SOUTH
Ancient Gneiss Complex
Pigg's Peak
South Africa
Swaziland
S26°00'
Transvaal and Karoo cover sequences
3.17 - 2.7 Ga intrusives
3.1 Ga GMS suite
Syenites
Composite batholiths Heerenveen
3.55 - 3.2 TTG basement plutons & gneisses
3.55 - 3.2 Ga Barberton greenstone belt
Barberton greenstone belt
South Africa
Swaziland

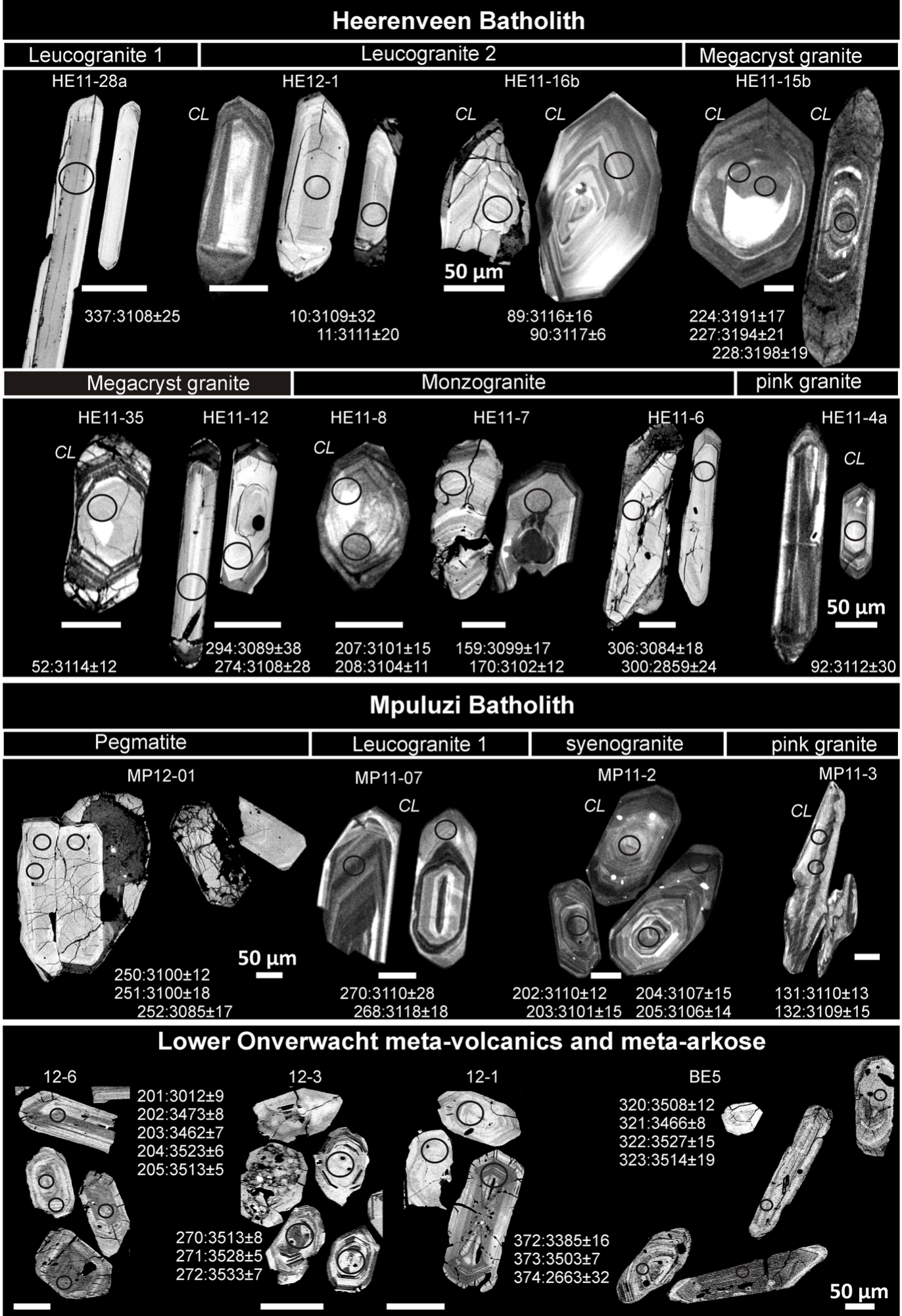

HEERENVEEN
IV
Pink granite II
Pink granite I
III
Quartz monzonite
Leucogranite II
II
Megacrystic granite
I
Leucogranite I
(50-80% intrusive sheets)
Leucogranite I
(0-50% intrusive sheets)
MPULUZI
IV or III
Leucogranite III
Leucogranite II
II
Megacrystic granite
I
Leucogranite I
Syenite
Younger cover sequence
TTG basement
Barberton Greenstone Belt
Synmagmatic shear zones
N
5 km
26°
26°10′
30°30′
30°40′
Badplaas
HE-11-35
HE-11-28A
HE-11-31A
HE-11-27B
HE-11-10
HE-11-15B
HE-11-12
HE-11-4A
HE-11-16B
HE-12-01
HE-11-8
HE-11-3
HE-11-6
HE-11-7
phon. (W04)
T (KD94)
KK12 (Z09)
MP-12-01
MP-11-3
MP-11-2
S (KD94)
RSA03 (M15)
MP-11-7
586 (W04)
RSA04 (M15)
RSA05 (M15)

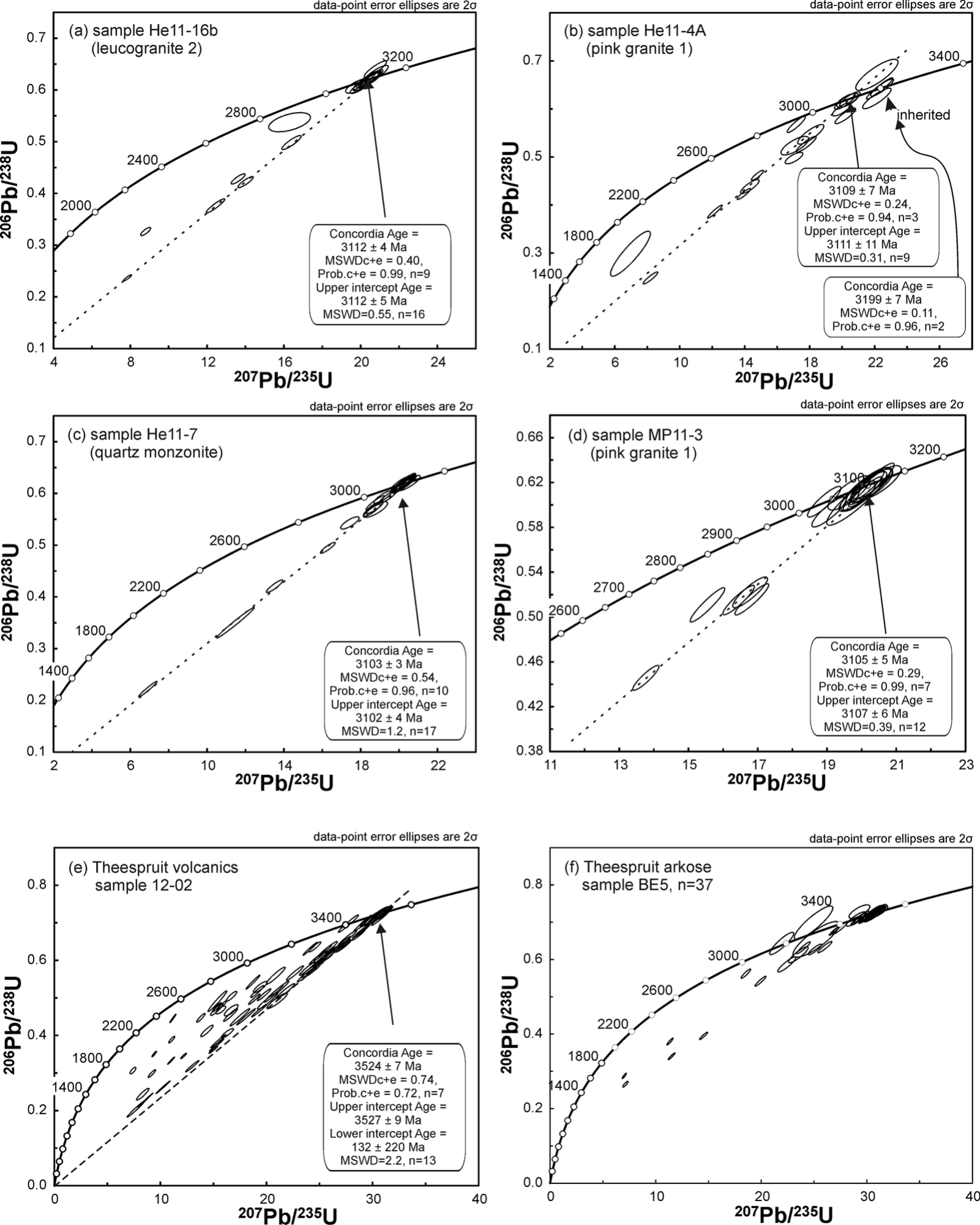
HEERENVEEN
MPULUZI
BOESMANSKOP
a.
b.
c.
d.
e.
f.
g.
SG
Peg
SG
Aug
Aug
SG
SG
h.
i.
j.
k.

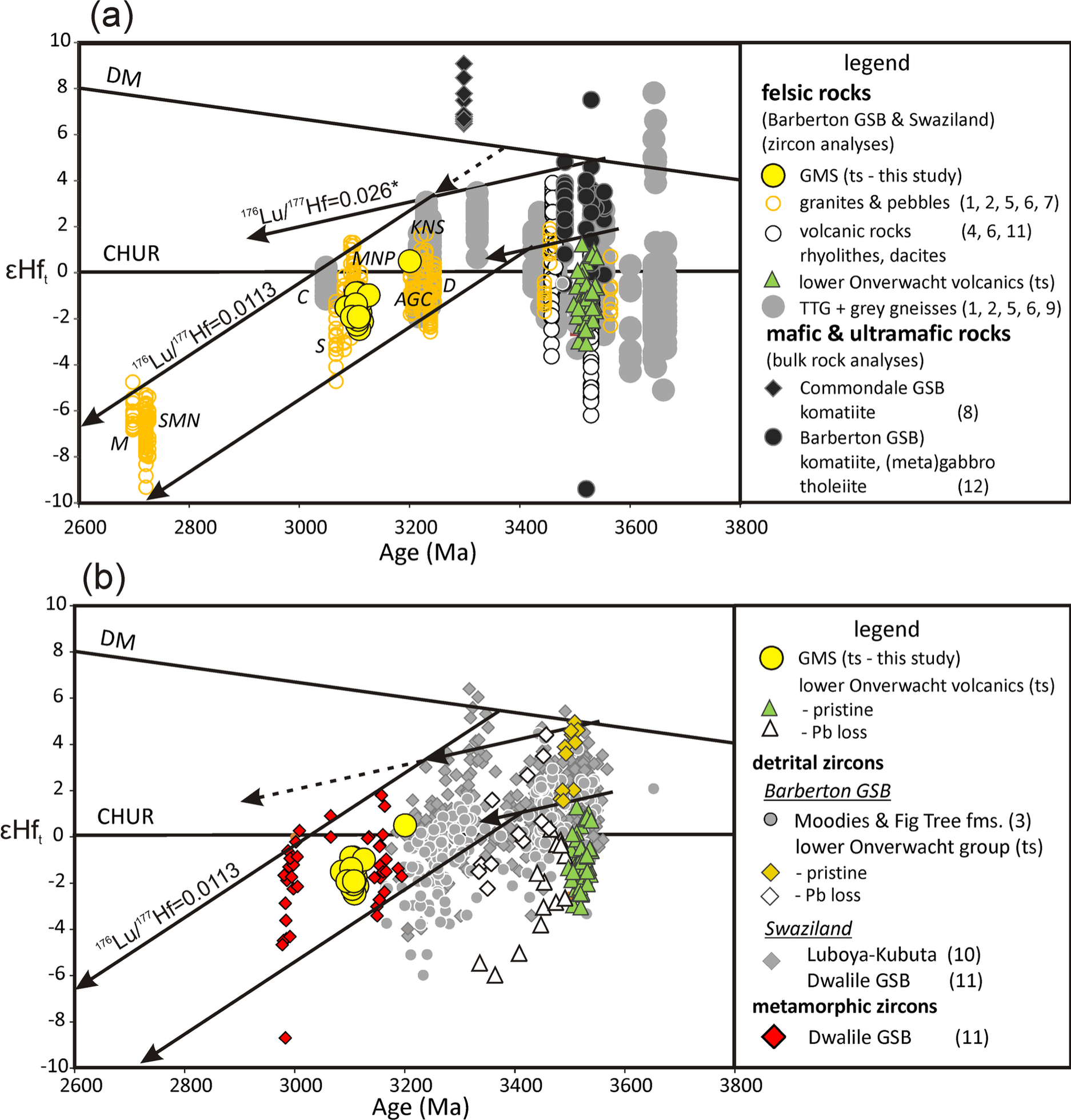

a.
Q = Si/3 − (K + Na + 2Ca/3)
P = K − (Na + Ca)
to
gd
gr
dq
mzdq
mzq
sq
mzgo
mz
s
b.
Na2O + K2O - CaO
SiO2
alkali−calcic
calc−alkalic
c.
A/NK
A/CNK
d.
A = Al − (K + Na + 2Ca)
B = Fe + Mg +Ti
f−P
h−P
Peraluminous
m−P
Metaluminous
e.
Mg/(Fe + Mg)
B = Fe + Mg + Ti
MAGNESIAN
FERRIFERROUS
f.
K2O
SiO2
High−K
Heerenveen
Monzonite and pink granites
Mpuluzi
Synenogranites and syenites
Syenites
Dark granites and granodiorite dykes (low K)
Dark granites and granodiorite dykes or small bodies
low K
medium K, low Ti
medium K, high Ti
Dark granites
Megacrystic granites
low K
medium K
(rare)
Megacrystic granites
high K
Leucogranites
low K
(mostly leuco 1)
medium K
(mostly leuco 2)
Leucogranites
medium K
(leuco 1)
high K
(leuco 2,3 & 4)
low-Ca granites
Boesmanskop
Syenites

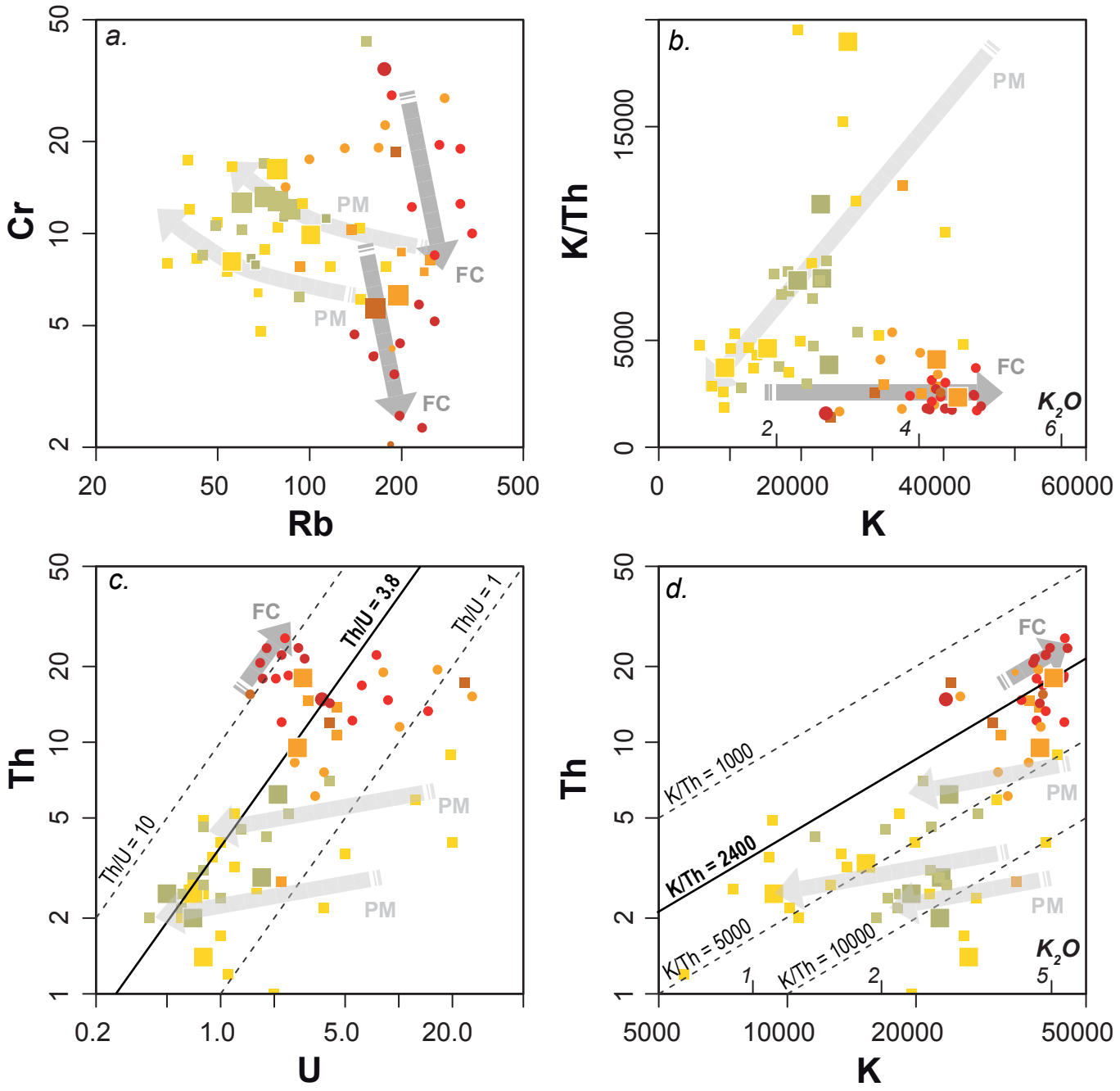

a. Low Ca Granites (low K)
b. Low Ca Granites (med K)
c. Low Ca Granites (high K)
d. Dark granites (low K)
e. Dark granites
f. Syenites and related
Rock / Average Barberton TTG
10
1
0.1
La Ce Pr Nd Sm Eu Gd Tb Dy Ho Er Lu Yb

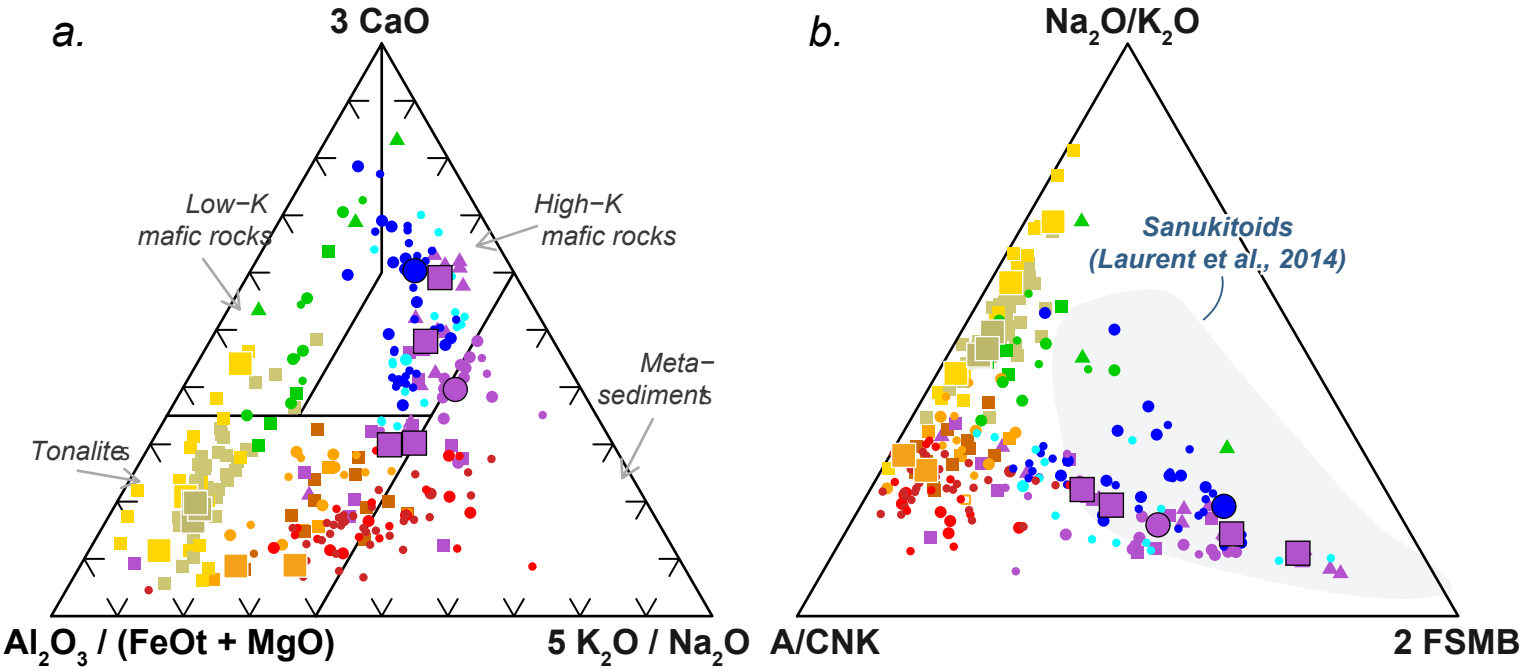

a. Low Ca Granites (low K)
b. Low Ca Granites (med K)
c. Low Ca Granites (high K)
d. Dark granites (low K)
e. Dark granites
f. Syenites and related
Rock / Average Barberton TTG
10
1
0.1
K Rb Ba Sr Zr Nb Ti Ce Sm Eu Th Y Yb

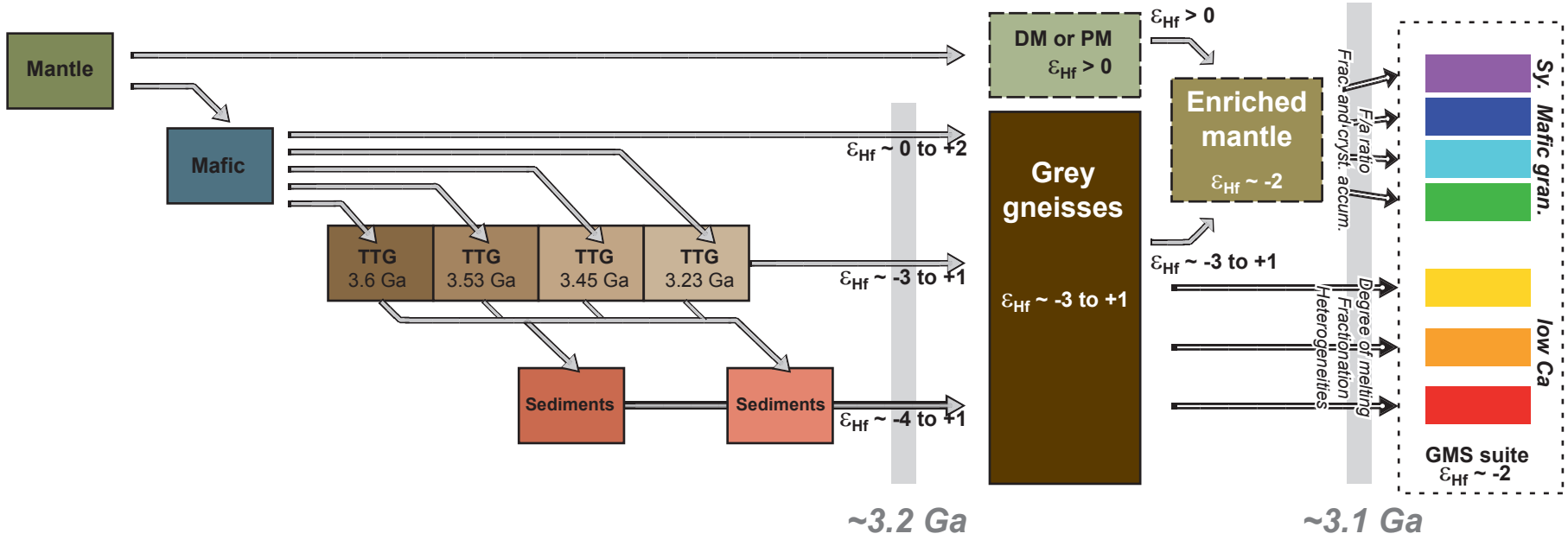

Heerenveen Batholith
Leucogranite 1
Leucogranite 2
Megacryst granite
HE11-28a
HE12-1
HE11-16b
HE11-15b
CL
50 µm
337:3108±25
10:3109±32
11:3111±20
89:3116±16
90:3117±6
224:3191±17
227:3194±21
228:3198±19
Megacryst granite
Monzogranite
pink granite
HE11-35
HE11-12
HE11-8
HE11-7
HE11-6
HE11-4a
52:3114±12
294:3089±38
274:3108±28
207:3101±15
208:3104±11
159:3099±17
170:3102±12
306:3084±18
300:2859±24
92:3112±30
Mpuluzi Batholith
Pegmatite
Leucogranite 1
syenogranite
pink granite
MP12-01
MP11-07
MP11-2
MP11-3
250:3100±12
251:3100±18
252:3085±17
270:3110±28
268:3118±18
202:3110±12
203:3101±15
204:3107±15
205:3106±14
131:3110±13
132:3109±15
Lower Onverwacht meta-volcanics and meta-arkose
12-6
12-3
12-1
BE5
201:3012±9
202:3473±8
203:3462±7
204:3523±6
205:3513±5
270:3513±8
271:3528±5
272:3533±7
372:3385±16
373:3503±7
374:2663±32
320:3508±12
321:3466±8
322:3527±15
323:3514±19

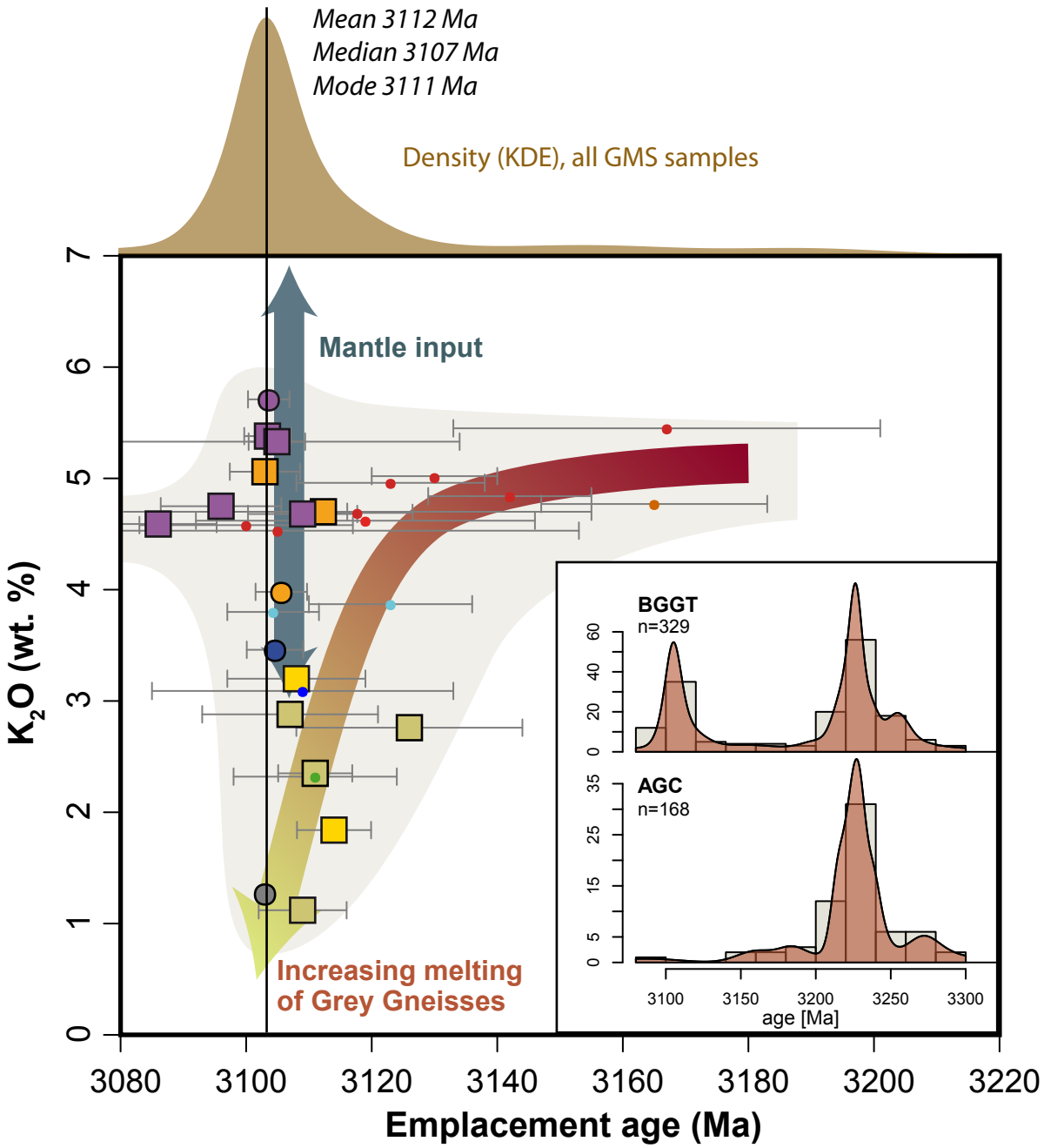

data-point error ellipses are 2σ
(a) sample He11-16b
(leucogranite 2)
Concordia Age =
3112 ± 4 Ma
MSWDc+e = 0.40,
Prob.c+e = 0.99, n=9
Upper intercept Age =
3112 ± 5 Ma
MSWD=0.55, n=16
(b) sample He11-4A
(pink granite 1)
inherited
Concordia Age =
3109 ± 7 Ma
MSWDc+e = 0.24,
Prob.c+e = 0.94, n=3
Upper intercept Age =
3111 ± 11 Ma
MSWD=0.31, n=9
Concordia Age =
3199 ± 7 Ma
MSWDc+e = 0.11,
Prob.c+e = 0.96, n=2
(c) sample He11-7
(quartz monzonite)
Concordia Age =
3103 ± 3 Ma
MSWDc+e = 0.54,
Prob.c+e = 0.96, n=10
Upper intercept Age =
3102 ± 4 Ma
MSWD=1.2, n=17
(d) sample MP11-3
(pink granite 1)
Concordia Age =
3105 ± 5 Ma
MSWDc+e = 0.29,
Prob.c+e = 0.99, n=7
Upper intercept Age =
3107 ± 6 Ma
MSWD=0.39, n=12
(e) Theespruit volcanics
sample 12-02
Concordia Age =
3524 ± 7 Ma
MSWDc+e = 0.74,
Prob.c+e = 0.72, n=7
Upper intercept Age =
3527 ± 9 Ma
Lower intercept Age =
132 ± 220 Ma
MSWD=2.2, n=13
(f) Theespruit arkose
sample BE5, n=37
$^{206}Pb/^{238}U$
$^{207}Pb/^{235}U$


(a)
DM
CHUR
εHf$_t$
^{176}Lu/^{177}Hf=0.026*
^{176}Lu/^{177}Hf=0.0113
KNS
MNP
AGC
C
D
S
SMN
M
Age (Ma)
legend
felsic rocks
(Barberton GSB & Swaziland)
(zircon analyses)
GMS (ts - this study)
granites & pebbles (1, 2, 5, 6, 7)
volcanic rocks (4, 6, 11)
rhyolithes, dacites
lower Onverwacht volcanics (ts)
TTG + grey gneisses (1, 2, 5, 6, 9)
mafic & ultramafic rocks
(bulk rock analyses)
Commondale GSB
komatiite (8)
Barberton GSB)
komatiite, (meta)gabbro
tholeiite (12)
(b)
DM
CHUR
εHf$_t$
^{176}Lu/^{177}Hf=0.0113
Age (Ma)
legend
GMS (ts - this study)
lower Onverwacht volcanics (ts)
- pristine
- Pb loss
detrital zircons
Barberton GSB
Moodies & Fig Tree fms. (3)
lower Onverwacht group (ts)
- pristine
- Pb loss
Swaziland
Luboya-Kubuta (10)
Dwalile GSB (11)
metamorphic zircons
Dwalile GSB (11)


a.
Cr
Rb
PM
FC
PM
FC
b.
K/Th
K
PM
FC
K_2O
2
4
6
c.
Th
U
FC
Th/U = 3.8
Th/U = 1
Th/U = 10
PM
PM
d.
Th
K
FC
K/Th = 1000
K/Th = 2400
K/Th = 5000
K/Th = 10000
PM
PM
K_2O
1
2
5


a.
3 CaO
Low−K
mafic rocks
High−K
mafic rocks
Meta−
sediments
Tonalites
Al2O3 / (FeOt + MgO)
5 K2O / Na2O
b.
Na2O/K2O
Sanukitoids
(Laurent et al., 2014)
A/CNK
2 FSMB


Mantle
Mafic
TTG 3.6 Ga
TTG 3.53 Ga
TTG 3.45 Ga
TTG 3.23 Ga
Sediments
Sediments
εHf ~ 0 to +2
εHf ~ -3 to +1
εHf ~ -4 to +1
DM or PM εHf > 0
Grey gneisses εHf ~ -3 to +1
εHf > 0
Enriched mantle εHf ~ -2
εHf ~ -3 to +1
Frac. and cryst. accum.
F/a ratio
Degree of melting
Fractionation
Heterogeneities
Sy. Mafic gran.
low Ca
GMS suite εHf ~ -2
~3.2 Ga
~3.1 Ga


Mean 3112 Ma
Median 3107 Ma
Mode 3111 Ma
Density (KDE), all GMS samples
Mantle input
Increasing melting
of Grey Gneisses
BGGT
n=329
AGC
n=168
age [Ma]
K_2O (wt. %)
Emplacement age (Ma)

Chemical type	Sub-type	Mpuluzi	Herenveen	Boesmanskop	Mg/Fe balance	Alkalinity	Aluminosity	Na/K balance
low-Ca granitoids	low K_2O	leucogranite I	Main megacrystic facies leucogranite I		Magnesian	calc-alk	felsic peraluminous	mostly medium K series
	medium K_2O		leucogranite II		Magnesian or Ferroan	alk-calc to calc-alk	felsic peraluminous	high K series
	high K_2O	leucogranites II, III and IV Main megacrystic facies			Ferroan	alk-calc	felsic peraluminous	high K to shoshonitic series
syenites and related		Syenite / Syenogranite	Monzonite Pink granite	Syenite	Ferroan	alk-calc to alk	strongly metaluminous	mostly shoshonitic series
dark granites	low K_2O	Some grey dykes; portions of main megacrystic facies (from literature - undescribed)	Grey granite & granodiorite dykes	Some "mafic" dykes	Magnesian	calc-alk	weakly peraluminous	medium K series
	medium K_2O / low TiO_2				Magnesian or Ferroan	alk-calc to calc-alk	weakly peraluminous	high K series
	medium K2O / high TiO_2	Eagle High "Augen granodiorite"			Ferroan	alk-calc to calc-alk	metaluminous	high K series

Order and values for REE spidergrams

La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Lu	Yb
17.60	29.84	3.19	12.24	2.37	0.71	1.71	0.27	1.75	0.35	0.96	0.12	0.69

Order and values for multi-elements diagrams

K	Rb	Ba	Sr	Zr	Nb	Ti	Ce	Sm	Eu	Th	Y	Yb
17993.96	58.00	344.53	526.93	120.28	5.50	1802.24	29.84	2.37	0.71	3.08	7.07	0.69

Table 3: Summary of U-Pb dating results

sample	Facies	Chemical type	Lat.	Long.	all n	Discordia AGE u.i.(Ma)	±2σ (Ma)[a]	AGE l.i.(Ma)	±2σ (Ma)	MSWD	Prob.	n	Concordia (99-101%) AGE conc.(Ma)	±2σ (Ma)[a]	MSWD c+e[b]	Prob. c+e[b]	n	comment / ref
Heerenveen																		
HE-11-28A	*Leucogranite*	*lowK Leucogr*	*-26.0580*	*30.5619*	11	3068	60	482	200	81*		11	3108	23		0.97	1	
HE-12-1	*Leucogranite*	*lowK Leucogr*	*-26.1329*	*30.4990*	20	3118	11	29	29	0.97	0.47	13	3114	12	0.54	0.80	4	
HE-11-27A	*Leucogranite*	*mediumK Leu*	*-26.0721*	*30.5574*	7								3103	11	0.23	1.00	6	
HE-11-16b	*Leucogranite*	*mediumK Leu*	*-26.1431*	*30.4721*	20	3112	9	-26	28	0.55	0.9	16	3112	8	0.40	0.99	9	
HE-11-15B	*Megacryst gra*	*lowK megacr.*	*-26.0869*	*30.4859*	17	3199	10	3	35	0.31	0.93	8	3201	9	0.22	0.99	5	(all zircons inherited)
HE-11-35	*Megacryst gra*	*lowK megacr.*	*-26.0264*	*30.5353*	19								3111	12	0.23	0.88	2	rited grains at 3.2 Ga
HE-11-12	*Megacryst gra*	*lowK megacr.*	*-26.0938*	*30.4380*	18	3124	15	58	41	0.53	0.75	7	3107	27		0.93	1	rited grains at 3.2 Ga
HE-11-10	*Megacryst gra*	*lowK megacr.*	*-26.0722*	*30.4236*	5								3126	37	0.06	0.81	1	
HE-11-8	*Quartz monzo*	*Sy, Mz, pink*	*-26.1586*	*30.4836*	24	3102	9	2	60	1.60	0.11	12	3105	9	0.13	1.00	4	
HE-11-7	*Quartz monzo*	*Sy, Mz, pink*	*-26.1993*	*30.4545*	21	3102	8	-31	70	1.20	0.24	17	3103	7	0.54	0.95	10	
HE-11-6	*Quartz monzo*	*Sy, Mz, pink*	*-26.1948*	*30.4542*	18	3079	24	207	68	7.1*		17	3086	19	0.28	0.60	1	
HE-11-3	*Pink granite 1*	*Sy, Mz, pink*	*-26.1674*	*30.4094*	16	3109	11	2.9	4	0.14	0.94	5						
HE-11-4A	*Pink granite 1*	*Sy, Mz, pink*	*-26.1277*	*30.3746*	20	3111	13	-13	62	0.31	0.95	9	3109	14	0.24	0.94	3	
						3199	13	265	270	0.19	0.83	4	3199	13	0.11	0.95	2	rited grains at 3.2 Ga
HE-11-31A	*Pink granite 2*	*Sy, Mz, pink*	*-26.0706*	*30.5588*	4								3096	19	0.92	0.47	3	
Mpuluzi																		
MP-12-01	*Pegmatite*		*-26.0957*	*30.6297*	12	3104	9	-584	170	0.57	0.78	9	3103	9	0.32	0.97	5	verse concordant
MP-11-7	*Leucogranite*	*mediumK Leu*	*-26.1458*	*30.6468*	23	3109	9	-80	360	0.84	0.63	16	3106	8	0.28	1.00	8	
MP-11-3	*Pink granite 1*	*Sy, Mz, pink*	*-26.0965*	*30.6312*	19	3107	10	133	230	0.39	0.95	12	3105	9	0.29	0.99	7	
MP-11-2	*Syenogranite*	*Sy, Mz, pink*	*-26.0963*	*30.6309*	21								3104	7	0.48	0.99	16	
S	*Leucogranite*	*probably high*	*-26.1465*	*30.7743*[x]									3107.0	4.0				Kamo and Davies 1994, TIMS
586	*Leucogranite*	*probably high*	*-26.1563*	*30.6065*									3105.6	2.4				Westraat et al. 2004, TIMS
Boesmanskop																		
unnamed	*Phonolite*	*Sy, Mz, pink*	*-26.0570*	*30.6571*									3096.0	4.0				Westraat et al. 2004, TIMS
T	*Syenite, Bk."t*	*Sy, Mz, pink*	*-26.0663*	*30.7324*[x]									3107.0	2.0				Kamo and Davies 1994, TIMS
Theespruit volcanics																		
.12-1	*Felsic schist*		*-26.0029*	*30.7893*	115	3521	10	-3	110	8.0*		13	3530	8	0.99	0.43	4	
.12-2	*Agglomerate*		*-26.0029*	*30.7893*	87	3527	9	132	220	2.2*		13	3524	7	0.74	0.72	7	
.12-3	*Conglomerate*		*-25.9960*	*30.7867*	52	3527	9	-6	30	3.2*		12	3527	8	0.22	0.88	2	
.12-6	*Felsic schist*		*-26.0144*	*30.7835*	66	3531	9	132	60	3.9*		23	3531	7	1.30	0.13	12	

u.i.-upper intercept, l.i.-lower intercept, Prob.-Probability

[x] approximate location, read from published map

[a] error without (and with) decay constant error

c+e[b]-MSWD and Probability of concordance and equivalence

*MSWD of discordia (reference) line

0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
Ethiopia
Kenya
Uganda
Zambia
Ethiopia
Kenya

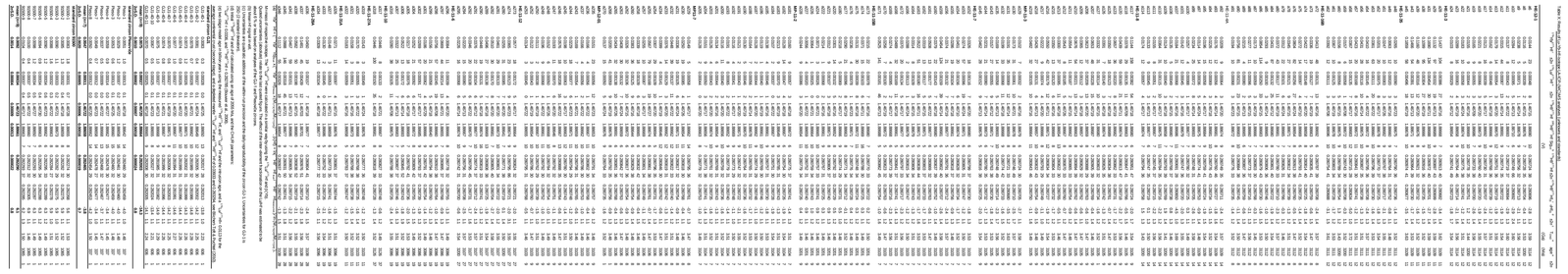
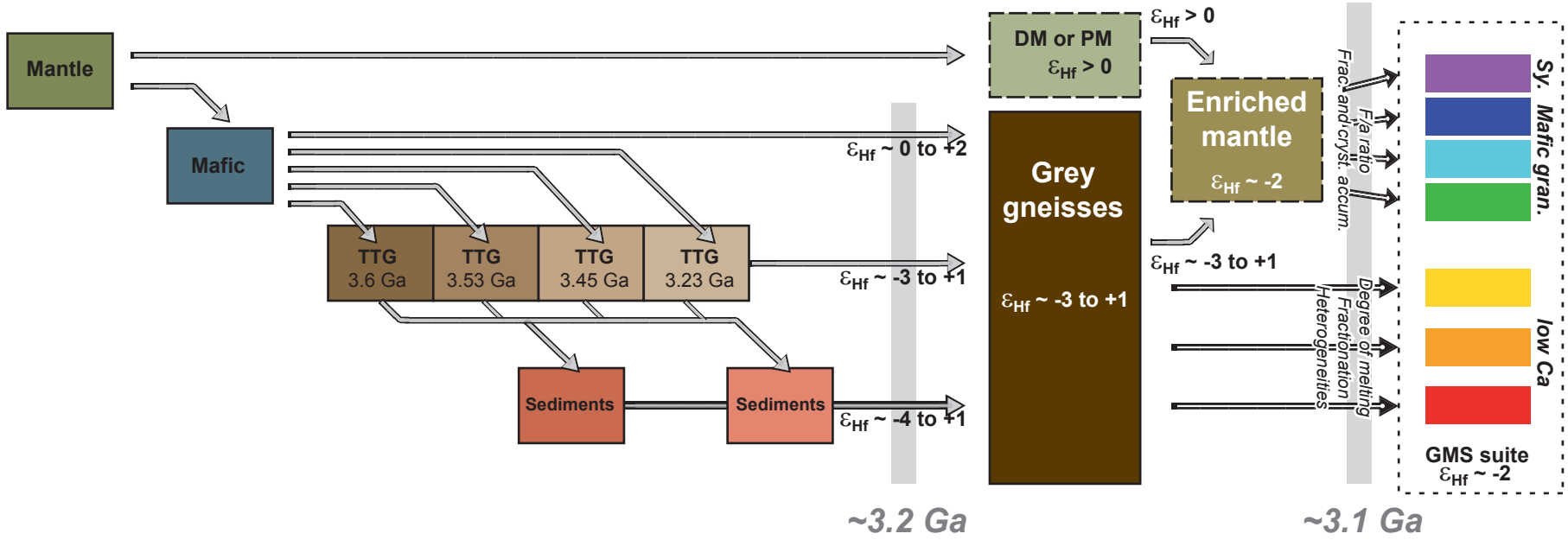


Table X: Summary of Lu-Hf isotope analyses

sample	Facies	$^{176}Hf/^{177}Hf_t$	±2σ (mean)	εHf_t	±2σ (mean)	T_{DM} (Ga)	±2σ (mean)	n	age (Ma)	comment
Heerenveen										
HE-11-28A	*Leucogranite 1*	0.28072	0.00003	-1.9	1.0	3.54	0.03	6	3108	
HE-12-1	*Leucogranite 2*	0.28071	0.00003	-2.1	1.1	3.56	0.03	13	3114	
HE-11-27A	*Leucogranite 2*	0.28074	0.00001	-1.4	0.5	3.51	0.01	4	3103	
HE-11-16b	*Leucogranite 2*	0.28074	0.00004	-1.3	1.4	3.51	0.04	16	3112	
HE-11-15B	*Megacryst granite*	0.28073	0.00002	0.5	0.6	3.48	0.02	13	3201	
HE-11-35	*Megacryst granite*	0.28073	0.00004	-1.8	1.3	3.54	0.04	10	3111	1 ol, (i.n.c.)
HE-11-12	*Megacryst granite*	0.28072	0.00002	-2.1	0.8	3.55	0.02	9	3107	(i.n.c.)
HE-11-10	*Megacryst granite*	0.28074	0.00003	-1.0	1.0	3.50	0.03	2	3126	
HE-11-8	*Quartz monzonite*	0.28075	0.00003	-1.1	1.2	3.50	0.03	19	3105	
HE-11-7	*Quartz monzonite*	0.28074	0.00003	-1.5	1.2	3.52	0.03	17	3103	
HE-11-6	*Quartz monzonite*	0.28075	0.00003	-1.5	0.9	3.50	0.03	11	3086	
HE-11-3	*Pink granite 1*	0.28071	0.00005	-2.5	1.7	3.57	0.05	7	3109	
HE-11-4A	*Pink granite 1*	0.28072	0.00003	-1.9	1.0	3.54	0.03	9	3109	1 ol, (i.n.c.)
HE-11-31A	*Pink granite 2*	0.28073	0.00003	-1.9	1.0	3.53	0.03	4	3096	
Mpuluzi										
MP-12-01	*Pegmatite*	0.28076	0.00002	-0.9	0.7	3.48	0.02	17	3103	
MP-11-7	*Leucogranite 1*	0.28075	0.00002	-0.9	0.8	3.48	0.02	16	3106	
MP-11-3	*Pink granite 1*	0.28072	0.00004	-2.3	1.3	3.56	0.04	8	3105	
MP-11-2	*Syenogranite*	0.28074	0.00002	-1.5	0.6	3.52	0.02	17	3104	
Theespruit volcanics										
.12-1	*Felsic agglomerate*	0.28046	0.00006	-1.4	2.2	3.83	0.12	7	3521	
.12-2	*Felsic agglomerate*	0.28048	0.00007	-0.6	2.4	3.80	0.13	20	3527	
.12-3	*Felsic agglomerate*	0.28047	0.00004	-1.2	1.3	3.83	0.07	3	3527	
.12-6	*Felsic agglomerate*	0.28047	0.00005	-1.0	1.9	3.82	0.10	35	3531	

ol - outlier, (i.n.c.) - inherited grains not considered


Mantle
Mafic
TTG 3.6 Ga
TTG 3.53 Ga
TTG 3.45 Ga
TTG 3.23 Ga
Sediments
Sediments
εHf ~ 0 to +2
εHf ~ -3 to +1
εHf ~ -4 to +1
DM or PM εHf > 0
Grey gneisses εHf ~ -3 to +1
εHf > 0
Enriched mantle εHf ~ -2
εHf ~ -3 to +1
Frac. and cryst. accum.
F/a ratio
Degree of melting
Fractionation
Heterogeneities
Sy. Mafic gran.
low Ca
GMS suite εHf ~ -2
~3.2 Ga
~3.1 Ga