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**Notes**

## U–Pb and Hf isotope data of detrital zircons from the Barberton Greenstone Belt: constraints on provenance and Archaean crustal evolution

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**Abstract:** Combined U–Pb and Hf isotope analyses of detrital zircons from the Fig Tree and Moodies Groups of the Barberton Greenstone Belt, South Africa, yield similar Hf isotope compositions and age populations, thus pointing to a similar provenance. Zircon populations of Fig Tree Group greywacke and Moodies Group quartzarenite are both dominated by age clusters at 3.53, 3.47, and 3.28 Ga, and a minor cluster at 3.36 Ga. The Moodies quartzarenite sample additionally contains a younger age population at 3.23–3.19 Ga. Hafnium isotope data indicate that the source area of both sediments was affected by new crust formation from depleted mantle sources at 3.53, 3.47, and perhaps at 3.36 Ga ( $\epsilon\text{Hf}_i$  between  $-1.7$  and  $+4.5$ ), accompanied by partial reworking of an Eoarchaean crustal component as old as 3.75–3.95 Ga. In contrast, crustal reworking was the predominant process between 3.28 and 3.22 Ga ( $\epsilon\text{Hf}_i$  between  $-6.0$  and  $+0.9$ ), probably related to subduction and collision of terranes along the Inyoka Fault system. The zircon U–Pb and Hf isotope datasets favour a southern provenance for the Fig Tree and Moodies sediments, comprising granitoids in the vicinity of the southern Barberton Greenstone Belt and in Swaziland. This finding is in contrast to the sedimentary record of the Moodies Group, which mostly suggests a northern and along-strike provenance. This discrepancy may be due to reworking of sediments during extensive syn- and postorogenic strike-slip faulting and high uplift or subsidence between 3.26 and 3.19 Ga.

**Supplementary material:** Results of *in situ* U–Pb and Lu–Hf isotope zircon analyses are available at [www.geolsoc.org.uk/SUP18561](http://www.geolsoc.org.uk/SUP18561).

The present-day geology of the Kaapvaal craton of southern Africa, one of the oldest continental nuclei worldwide, comprises several crustal blocks that are separated from each other by major lithospheric suture zones. These zones, which comprise the Inyoka Fault system of the Barberton Greenstone Belt and the Murchison–Tabazimbi Lineament of the Northern Kaapvaal Craton (Fig. 1), are interpreted as important terrane boundaries that formed during accretion of the Swaziland, Witwatersrand, and Pietersburg blocks (e.g. de Wit *et al.* 1992; Lowe & Byerly 1999; Poujol *et al.* 2003; Anhaeusser 2006; Schoene *et al.* 2009; Zeh *et al.* 2009). The Inyoka Fault system of the Barberton Greenstone Belt is considered to have originated from a *c.* 3.3–3.2 Ga subduction–accretion event that caused syntectonic magmatism, syn-contractual strike-slip basin development, and high-grade metamorphism (Kröner *et al.* 1991; de Wit *et al.* 1992; de Ronde & de Wit 1994; Kamo & Davis 1994; Lowe 1994; Lowe & Byerly 1999; de Ronde & Kamo 2000; Dziggel *et al.* 2002; Stevens *et al.* 2002; Diener *et al.* 2005; Moyon *et al.* 2006; Schoene *et al.* 2008, 2009; Zeh *et al.* 2009; Schoene & Bowring 2010), followed by transform boundary deformation that culminated in *c.* 3.1 Ga granitic magmatism and differential exhumation of mid-crustal rocks on extensionally reactivated faults (Westraat *et al.* 2005; Schoene & Bowring 2007, 2010; Schoene *et al.* 2008). The subduction–accretion event ended in the collision of two major terranes (or terrane collages), which are characterized by distinct granitoid intrusion ages and isotope patterns. The Barberton South Terrane (definition of Zeh *et al.* 2009), comprising the Stolzburg, Steynsdorp and Swaziland subterrane (including large parts of the Onverwacht Group and Ancient Gneiss Complex, Fig. 1), is made up of granitoids with ages of 3.66, 3.55, 3.45, 3.33 and 3.23 Ga (e.g. Compston & Kröner 1988; Armstrong *et al.* 1990;

Kröner *et al.* 1991; Kamo & Davis 1994; Kröner & Tegtmeier 1994; Schoene & Bowring 2007, 2010; Schoene *et al.* 2008; Zeh *et al.* 2009, 2011), whereas granitoids of the Barberton North Terrane are younger than 3.30 Ga (e.g. Kamo & Davis 1994; Poujol *et al.* 2003; Schoene *et al.* 2008; Zeh *et al.* 2009). In addition, the *c.* 3.23 Ga granitoids of the Barberton South Terrane always show near-chondritic  $\epsilon\text{Hf}_i$  and  $\epsilon\text{Nd}_i$ , indicating intense crustal reworking (Schoene *et al.* 2009; Zeh *et al.* 2009, 2011) whereas those of the Barberton North Terrane (e.g. Kaap Valley and Nelshoogte Plutons) have superchondritic  $\epsilon\text{Hf}_i$  and  $\epsilon\text{Nd}_i$  values, indicating derivation from a relatively juvenile source at the same time (Schoene *et al.* 2009; Zeh *et al.* 2009). Because of these differences provenance studies using combined U–Pb and Lu–Hf isotope data of detrital zircon grains have a high potential to provide robust information on the source area of sediments, which were deposited on either side of the Inyoka Fault during the *c.* 3.3–3.2 Ga subduction–accretion phase (i.e. the Fig Tree and Moodies Group metasandstones). This approach has been shown to be useful to obtain detailed information not only about the timing of magmatic and metamorphic processes in the source area but also to distinguish between mantle- and crust-related magma sources, the maximum age of deposition, and post-depositional metamorphic alteration (e.g. Gerdes & Zeh 2006, 2009; Zeh *et al.* 2008; Kaur *et al.* 2011). Furthermore, combined U–Pb and Lu–Hf isotope data can provide valuable information about the early Earth's crust–mantle evolution prior to the stabilization of the oldest cratons, as Archaean sediments often contain material that is older than the underlying cratonic crust. This, for example, is well reflected by the finding of Hadean and Eoarchaean zircons in metasediments of the Jack Hills and the Limpopo Belt, respectively (e.g. Maas *et al.* 1992; Zeh *et al.* 2008).

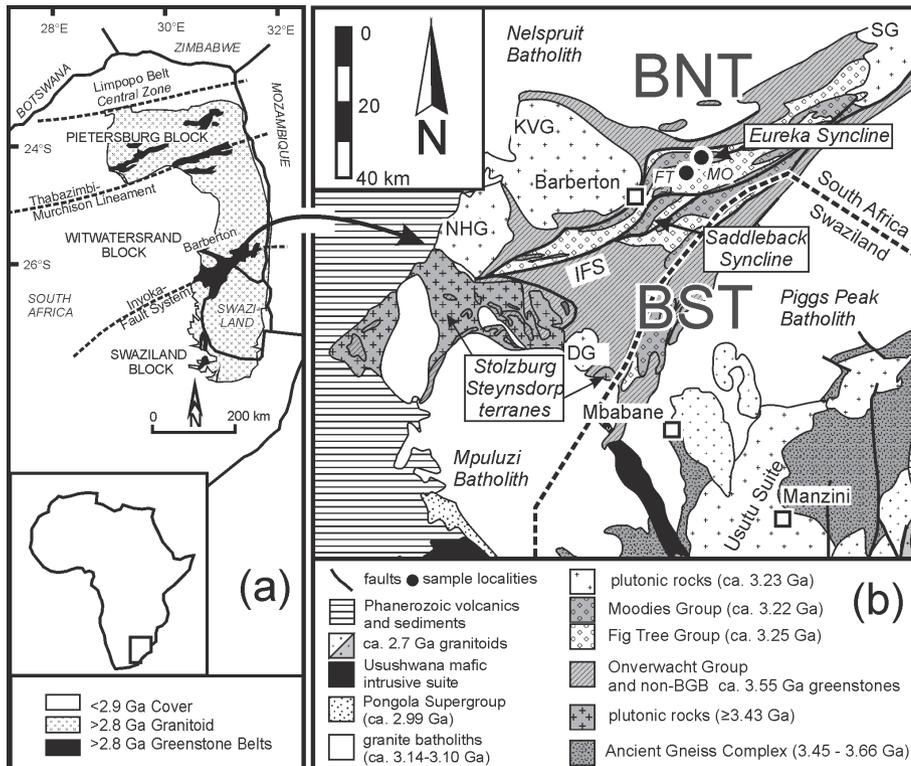
On a regional scale, detrital zircon data contribute an independent dataset to traditional sedimentary provenance indicators such as palaeocurrents, stratigraphic thickness or lithological variations, or textural data. In the Barberton Greenstone Belt, such tests are particularly required because currently available sedimentary and geochronological data are largely inconclusive. A limited set of U–Pb zircon ages of 3.4–3.5 Ga from Moodies Group conglomerates suggests a southern provenance (e.g. from the Onverwacht Group and/or the Ancient Gneiss Complex of Swaziland; Tegtmeier & Kröner 1987; Kröner & Compston 1988), whereas sedimentary, petrological and structural data instead indicate a significant input from the north (Heubeck & Lowe 1994a; Heubeck & Lowe 1999). To address this controversy, to constrain sedimentary provenance and to define crustal makeup of the source area, we obtained combined U–Pb and Lu–Hf isotope data from detrital zircons from a representative metamorphosed Fig Tree Group litharenite and a Moodies Group quartzarenite.

### Geological setting and samples

The Fig Tree and Moodies Group form the uppermost units of the Barberton Supergroup, the stratigraphic fill of the Barberton Greenstone Belt. The Fig Tree Group overlies the Onverwacht Group mostly conformably and consists of a southern shallow-water facies and a northern deep-water facies, separated by the Inyoka Fault system (Eriksson 1980; de Wit 1982; Lamb & Paris 1988). North of the Inyoka Fault, the Fig Tree Group traditionally consists of three formations, which are (from base to top) the Sheba, Belvue Road and the Schoongezicht Formation (Reimer 1967; Condie *et al.* 1970). The Sheba Formation comprises 750–1200 m of turbiditic lithic greywackes and shales, the Belvue Road Formation 600–1150 m of shale, turbiditic siltstone and greywacke, cherts, and locally volcanoclastic rocks and altered komatiitic lavas capped by a black shale near the top, and the Schoongezicht

Formation, c. 550 m of dacitic volcanic rocks, coarse-grained felsic volcanoclastic sandstones, conglomeratic breccias, tuffs, mudstones and some shale. Reimer (1983) additionally differentiated the Ulundi Formation, a 25–30 m thick iron-rich shale and chert unit, and Köhler & Anhaeusser (2002) recognized the 700–3000 m thick Bien Venue Formation, which mainly consists of aluminous quartz–muscovite schists and is apparently restricted to the north-eastern Barberton Greenstone Belt. The latter formation is derived from a sequence of quartz-phyric dacitic to rhyodacitic volcanoclastic protoliths, dated at c. 3.256–3.259 Ga (Kröner *et al.* 1991), associated with subordinate banded chert, phyllite and chlorite schist, and overlies sediments correlated with the Belvue Road Formation. It is in turn discordantly overlain by Moodies Group conglomerates and sandstones. Units assigned to the Fig Tree Group south of the Inyoka Fault include the Mapepe and Auber Villiers formations, which were deposited between 3259 and 3225 Ma, based on dating of dacitic tuffs and agglomerates (e.g. Kröner *et al.* 1991; Byerly *et al.* 1996; Lowe & Byerly 1999; Lowe & Nocita 1999). Zircons from dacitic tuffs and agglomerates from the stratigraphically lowest and uppermost parts of the Fig Tree Group north of the Inyoka Fault are dated at  $3.259 \pm 0.003$  Ga and  $3.225 \pm 0.003$  Ga, respectively (Kröner *et al.* 1991; Byerly *et al.* 1996); xenocrysts from these rocks yielded ages between 3.522 and 3.323 Ga. SHRIMP analyses of 14 zircons from a Fig Tree Group greywacke from Swaziland (south to the Inyoka Fault) gave uniform ages of c. 3455 Ma (Kröner & Compston 1988).

The Moodies Group is the uppermost lithostratigraphic unit of the Barberton Greenstone Belt, and mostly occupies large synclines. The greatest stratigraphic thicknesses, highest petrographic variability and most diverse sedimentary facies are preserved in the synclines north of the Inyoka Fault. Anhaeusser (1976) subdivided the Moodies Group in the Eureka Syncline into three formations, including (from base to top) the Clutha, Joe's Luck and Baviaanskop Formation. Each of these formations is defined as an upward-fining



**Fig. 1.** (a) Archaean stratigraphic units of the eastern part of the Kaapvaal Craton in southern Africa. The crustal blocks are separated by major lineaments or fault systems. (b) Simplified geological map of the Barberton Greenstone Belt and the northern part of Swaziland (modified after Schoone *et al.* 2009). BNT, Barberton North Terrane; BST, Barberton South Terrane; KVG, Kaap Valley Granite; NHG, Nelshoogte Granite; DG, Dalmein Granite; SG, Salisbury Kop Granite; IFS, Inyoka Fault System.

cycle consisting of a coarse basal unit of conglomeratic quartzose sandstones, followed by a thick succession of finer-grained sandstones, siltstones and shales, and usually terminating with some jaspilitic banded iron formation. However, this first-order lithostratigraphic framework can only partially be correlated with the thicker stratigraphic column of Moodies strata in the Saddleback Syncline or with the more distal Moodies Hills Block and Stolzberg Syncline. Eriksson (1979), without defining a type location, recognized five major units (MD1–MD5) in the Eureka and Saddleback Synclines.

Moodies strata north of the Inyoka Fault record a range of terrestrial and shallow-marine environments, including alluvial, fluvial–deltaic, shoreline, tidal, and shallow–middle shelf deposits (Eriksson 1979, 1980; Heubeck & Lowe 1994a). They are up to 3000 m in thickness, extend over a strike length of >60 km and are correlatable to some degree through a few marker units within the dominantly sandy lithologies. In contrast, Moodies Group strata south of the Inyoka Fault are far thinner (not more than several hundred metres in thickness), lack marker horizons and do not contain K-feldspar. Heubeck & Lowe (1999) argued that Moodies strata on either side of the Inyoka Fault were deposited diachronously and in separate fault-bounded basins. Facies and petrographic trends have been investigated only in the thick and diverse stratigraphic columns north of the Inyoka Fault; there, they represent a gradual transgressive trend from alluvial to shelfal facies, accompanied by increasing compositional maturity in sandstones and conglomerates, overlain by a shallowing-upwards trend represented by alluvial fan-deltas and reflecting abrupt decrease in compositional maturity. Heubeck & Lowe (1994a) interpreted the former (Oosterbeek Petrofacies of Heubeck & Lowe 1999) as a passive-margin-related or rift-related sequence trend and the overlying section (Elephant's Head Petrofacies of Heubeck & Lowe 1999) as a response to syndepositional up-to-the-north faulting along the northern Barberton Greenstone Belt margin, incipient basin inversion and greenstone belt shortening.

The start of Moodies Group deposition is well constrained by age data from felsic and dacitic volcanic rocks of the Schoongezicht Formation at the top of the Fig Tree Group, conformably underlying the Moodies Group. These volcanic rocks yielded ages of  $3.224 \pm 0.006$  Ga (Kröner & Todt 1988),  $3.225 \pm 0.003$  Ga (Kröner *et al.* 1991), and  $3.226 \pm 0.001$  Ga (Kamo & Davis 1994). In contrast, the end of Moodies deposition is less well constrained. The postkinematic, granitic Salisbury Kop Pluton of the northeastern Barberton Greenstone Belt unambiguously crosscuts the greenstone belt fold fabric, including tightly folded Moodies strata. It has been dated at  $3.079 \pm 0.016$  Ga (Heubeck *et al.* 1993),  $3.109 +0.010/-0.008$  Ga (Kamo & Davis 1994), and  $3.100 \pm 0.014$  Ga (Zeh *et al.* 2009). Also, deformed Moodies strata were magnetically overprinted by the Kaap Valley Pluton at  $3.214 \pm 0.004$  Ga, which is the Ar–Ar hornblende cooling age of the Kaap Valley Pluton (Layer *et al.* 1996). Single-grain zircon ages from thin felsic ash units within the upper Moodies Group in the Saddleback Syncline have yielded ages of  $3231 \pm 6$  Ma (Heubeck *et al.* 2010).

Granitoid pebbles from the Moodies basal conglomerate of the Eureka Syncline yield a complex spectrum. Pebbles dated by Tegtmeier & Kröner (1987), Kröner & Compston (1988) and Sanchez-Garrido *et al.* (2011) yield ages of *c.* 3.57 and *c.* 3.47 Ga. In addition, Sanchez-Garrido *et al.* (2011) identified younger pebbles with ages between 3.30 and 3.21 Ga (three out of 14 clasts). Although pebbles with ages of 3.57 and 3.47 Ga clearly point to a southern source area, south of the Inyoka Fault system, the younger pebbles may be derived from magmatic rocks on either side of the Inyoka Fault. Thus the age pattern of the granitoid clasts in the basal member of the Moodies Group indicates mostly but not

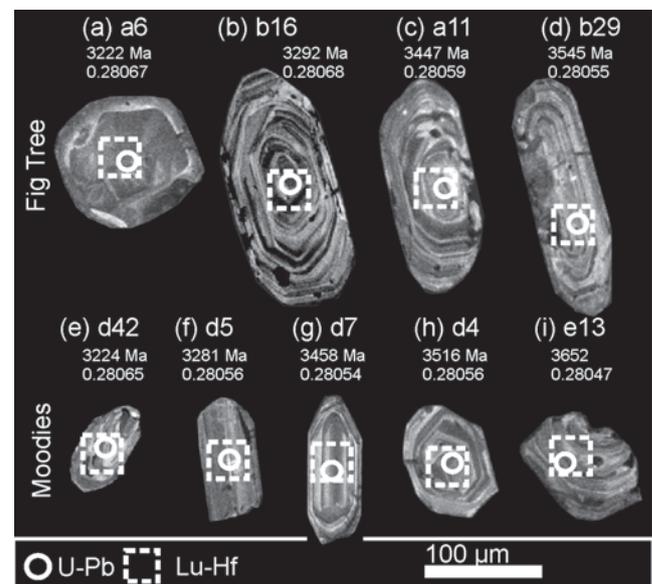
necessarily exclusively a southern provenance, and thus appears to support the tectonic scenarios of Eriksson (1979, 1980) and Jackson *et al.* (1987). In contrast, stratigraphic data of Heubeck & Lowe (1994a) suggest significant transport of Moodies material from the north and along strike. In addition, Heubeck & Lowe (1999) argued that Moodies strata south of the Inyoka Fault system have a greatly reduced thickness and show a fundamentally different petrographic composition.

The depositional setting of the Moodies sediments was originally considered to be a foreland basin (e.g. Jackson *et al.* 1987; Eriksson 1980), including alluvial fans, braided streams, delta plains, shallow-water coastal systems and shelf facies (Eriksson 1980; Heubeck & Lowe 1994a, b). More differentiated assignments based on detailed field work (Heubeck & Lowe 1994a) suggested that the basal conglomerates and the overlying fluvial sandstones were derived from the north. Shoreline-parallel, NE- or SW-directed transport and extensive reworking dominated overlying deltaic, tidal and marine facies. Northerly derived, southward-thinning fan-delta conglomerate(s) over progressive unconformities in the Joe's Luck Formation of the Moodies Group are interpreted to mark the beginning of basin shortening (Heubeck & Lowe 1994b).

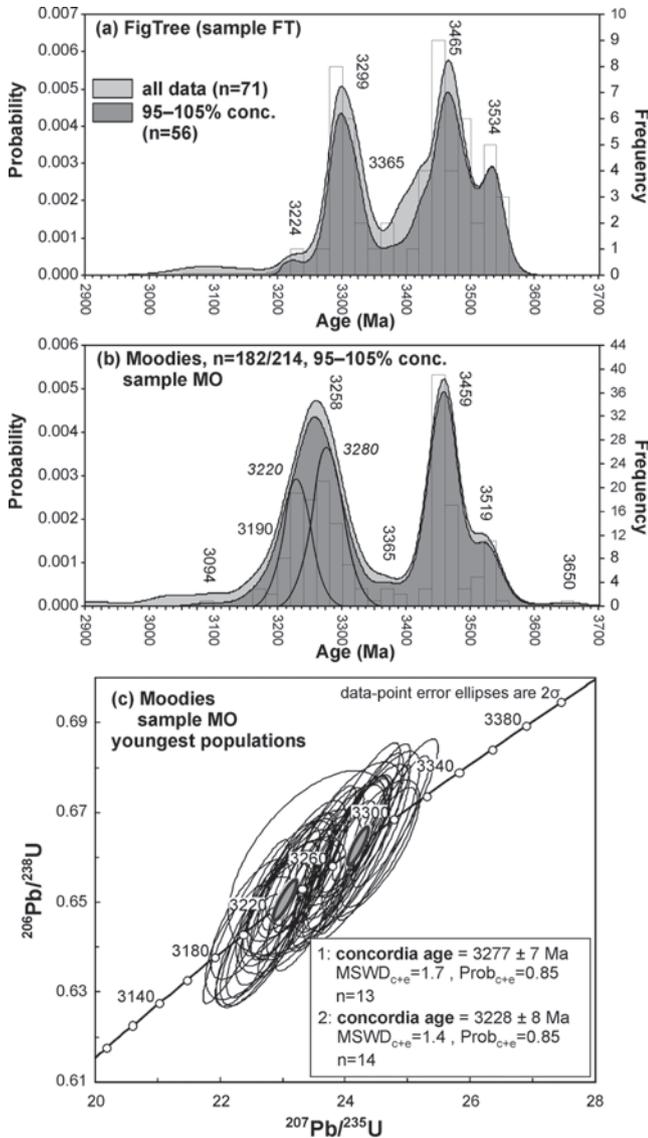
The samples used in this study were taken from the eastern Eureka Syncline at two roadcuts along the Sheba Mine access road. Sample FT is a coarse-grained greywacke from the Sheba Formation of the Fig Tree Group ( $25^{\circ}42'765''S$ ,  $31^{\circ}09'478''E$ ; altitude 660 m); sample MO is a well-sorted, medium-grained, horizontally stratified and cross-bedded quartzarenite from unit MdS1 (Clutha Formation of Anhaeusser 1976) of the Moodies Group ( $25^{\circ}41'429''S$ ,  $31^{\circ}10'484''E$ ; altitude 560 m).

## Results of U–Th–Pb and Lu–Hf isotope analyses

Analyses of the U–Th–Pb and Lu–Hf isotope systems were carried out by laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) at the Goethe University of Frankfurt, using the methods described by Gerdes & Zeh (2006, 2009), with some modifications. A Resolution M50 LR 193 nm Excimer (Resonetics)

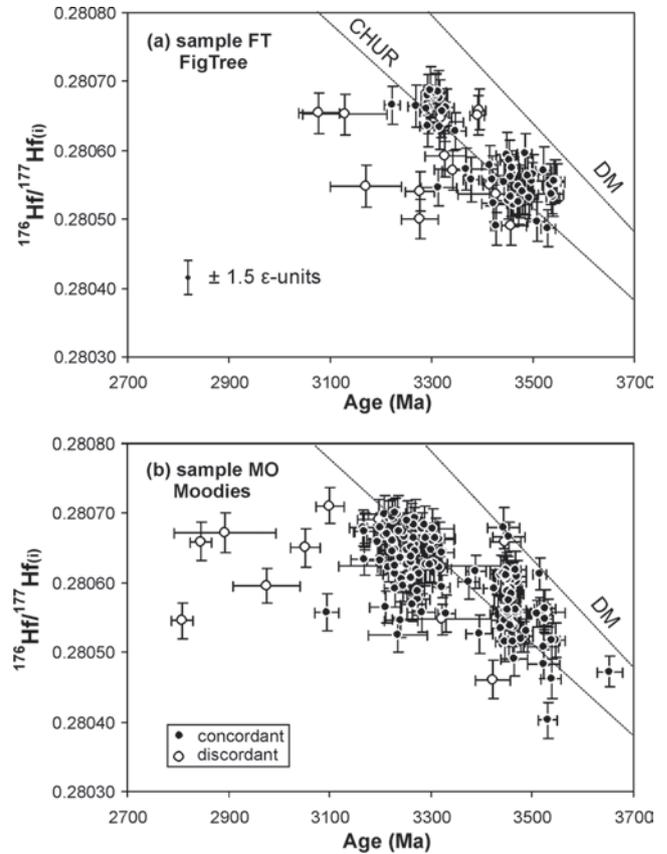


**Fig. 2.** CL images of detrital zircon grains from the Fig Tree Group greywacke and Moodies Group quartzarenite of the Eureka Syncline, with U–Pb and Lu–Hf laser spots.



**Fig. 3.** Results of U–Pb analyses for samples FT (a) and MO (b, c) represented in probability/frequency v. age diagrams. Vertical numbers show ages of zircon populations obtained by the software AgeDisplay. Horizontal numbers mark age populations not recognized by AgeDisplay. (c) Concordia diagram showing U–Pb ages obtained by pooling the youngest major zircon population from sample MO into two groups.

laser system was used for sample ablation; U–Th–Pb and Lu–Hf isotope analyses were performed using a single-collector (Element 2) and multi-collector (Neptune) sector field ICP-MS system, respectively. A detailed description of the method is given in the Appendix. During this study we carried out U–Th–Pb analyses on 71 zircons of sample FT, and on 214 zircons of sample MO. Subsequently, Lu–Hf isotope analyses were obtained from the same zircons by placing the laser spots directly on top of or next to the U–Th–Pb spots. Laser spot locations were selected on the basis of the internal structures of the grains as seen in cathodoluminescence (CL) images of the mounted and polished grains (Fig. 2). The CL images show oscillatory zoning for most zircon grains, typical for zircon growth in igneous rocks (Fig. 2); however, some zircons show core–rim relationships. For this study, only zircon cores were

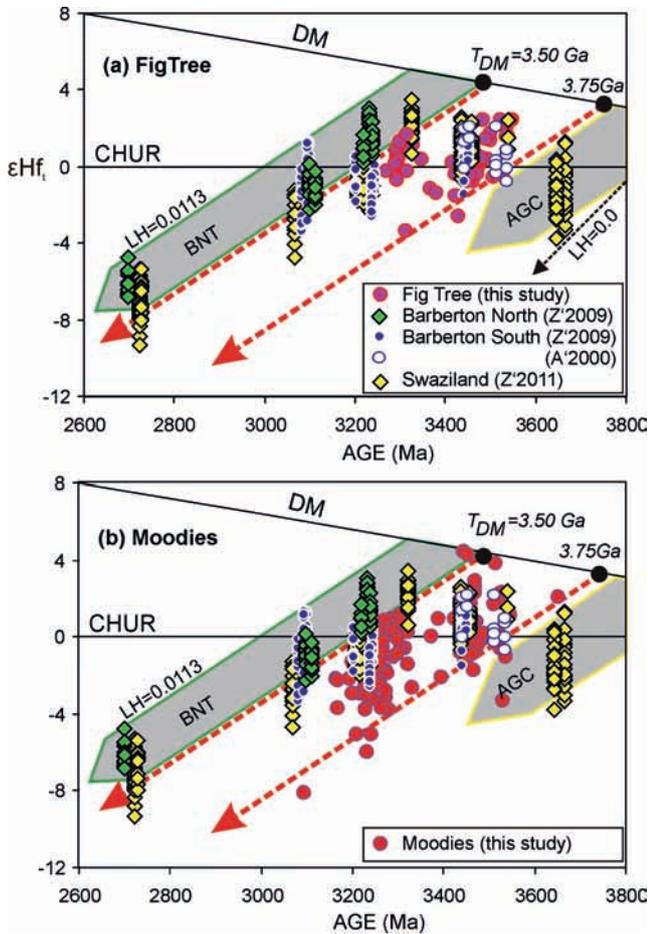


**Fig. 4.**  $^{176}\text{Hf}/^{177}\text{Hf}$  v. Pb–Pb age diagram showing the results of combined U–Pb and Hf isotope analyses, for concordant and discordant analyses of sample FT (a) and sample MO (b). CHUR, chondritic uniform reservoir; DM, depleted mantle.

analysed. The  $^{206}\text{Pb}/^{207}\text{Pb}$  age uncertainty of the single analyses is commonly less than  $\pm 0.02$  Ga (2SD). Age spectra (Fig. 3) were plotted using the software AgeDisplay (Sircombe 2004) and include  $^{206}\text{Pb}/^{207}\text{Pb}$  ages of all zircon analyses with a concordance level between 95 and 105%.

The U–Pb isotope data obtained from zircons of both samples show overall similar bimodal age spectra with predominant age peaks at 3.46 and 3.29–3.26 Ga. Subordinate populations occur at 3.54–3.52 and 3.37 Ga, and a single zircon of sample MO yielded 3.65 Ga (Fig. 3a and b). The youngest major age group of sample FT is relatively homogeneous, showing only small variations centred at  $3.29 \pm 0.02$  Ga. Only one analysis in sample FT (grain a6) reveals a significantly younger age at  $3.222 \pm 0.017$  Ga. In contrast, the youngest major age population of sample MO shows a much wider age variation ( $3.26 \pm 0.05$  Ga), as resolved by AgeDisplay, well outside the  $^{206}\text{Pb}/^{207}\text{Pb}$  age error of the single analyses. This may indicate that the population comprises two distinct populations of  $3.28 \pm 0.02$  and  $3.22 \pm 0.02$  Ga (Fig. 3b). This is in agreement with calculated U–Pb concordia ages of  $3.277 \pm 0.007$  Ga ( $n = 13$ ) and  $3.228 \pm 0.008$  Ga ( $n = 14$ ), respectively (Fig. 3c). In addition, there are a few zircon analyses in sample MO that yielded significantly younger ages. Four analyses cluster at about  $3.19 \pm 0.02$  Ga, and a single grain gave a concordia age of  $3.095 \pm 0.024$  Ga (Fig. 3a).

Hafnium isotope data of the zircons from both samples show many similarities (Fig. 4). Zircons with ages  $>3.365$  Ga have mostly superchondritic composition, whereas zircons with ages  $<3.33$  Ga have subchondritic composition (Fig. 4). The Hf isotope



**Fig. 5.** Synopsis of  $\epsilon\text{Hf}_i$  data of detrital zircon grains from the (a) Fig Tree Group and (b) Moodies Group (only concordant analyses) and from zircons of granitoids from the Barberton North and Barberton South terranes and Swaziland (data sources: A'2000, Amelin *et al.* 2000; Z'2009, Z'2011, Zeh *et al.* 2009, 2011). It should be noted that detrital zircons overlap well with data from the Barberton South Terrane and plot between the crustal evolution trends of the Ancient Gneiss Complex (AGC trend) and the Barberton North Terrane (BNT trend). LH =  $^{176}\text{Lu}/^{177}\text{Hf}$ . CHUR, chondritic uniform reservoir; DM, depleted mantle;  $T_{DM}$ , hafnium model ages.

variations extend to 8 epsilon units within the respective age population (Fig. 5). In general,  $\epsilon\text{Hf}_i$  increases towards older ages, on average from  $-1.3$  at  $3.22$  Ga to  $+1.6$  at  $3.53$  Ga in sample FT, and from  $-1.8$  at  $3.23$  Ga to  $+1.0$  at  $3.52$  Ga in sample MO (Table 1).

**Discussion**

*Provenance and depositional age*

Age spectra and Hf isotope data provide evidence that both the Fig Tree Group greywacke (Sheba Formation) and Moodies Group quartzarenite (Clutha Formation) from the Eureka Syncline were derived from one or several similar source region(s) that experienced magmatic activity at  $3.53 \pm 0.2$ ,  $3.46 \pm 0.2$ ,  $3.37 \pm 0.2$  and  $3.29 \pm 0.2$  Ga (Fig. 3). The ages of the oldest detrital zircons from both samples ( $3.53 \pm 0.2$  and  $3.46 \pm 0.2$  Ga) are in good agreement with intrusion ages of granitoids exposed to the south of the Inyoka Fault in the Barberton South Terrane (Fig. 1). The  $3.53$  Ga ages overlap with emplacement ages of the Steynsdorp tonalite, with tonalite wedges in the lower Onverwacht Group, with the age of Sandspruit Formation metavolcanic rocks, and with tonalite–trondhjemite–granodiorites (TTGs) of the Ancient Gneiss Complex (Compston & Kröner 1988; Kröner *et al.* 1989; Armstrong *et al.* 1990; Kamo & Davis 1994; Kröner & Tegtmeier 1994; Amelin *et al.* 2000; Schoene *et al.* 2008; Zeh *et al.* 2009, 2011). In addition, they coincide with the oldest group of zircon ages from granophyric clasts from the basal Moodies conglomerate (Tegtmeier & Kröner 1987; Kröner & Compston 1988; Sanchez-Garrido *et al.* 2011). Granitoids of the Ancient Gneiss Complex may be a potential source for the oldest zircon, with an age of  $3.65$  Ga, found in sample MO (Compston & Kröner 1988; Schoene *et al.* 2008; Zeh *et al.* 2011).

The abundant  $3.46$  Ga ages overlap with intrusion ages of plutonic rocks in the southwestern Barberton Greenstone Belt (e.g. Stolzburg and Theespruit Plutons), the intrusion age of the dacitic volcanic complexes at the top of the Kromberg Formation, and the Tsawela gneisses of Swaziland (Kröner *et al.* 1989; Armstrong *et al.* 1990; Kamo & Davis 1994; Byerly *et al.* 1996; Amelin *et al.* 2000; Schoene *et al.* 2008; Zeh *et al.* 2009, 2011). Because magmatic rocks with ages  $>3.4$  Ga are unknown north of the Inyoka Fault, all aforementioned detrital zircons must have been supplied from a southern source.

Detrital zircons in both samples also record age populations of  $3.29 \pm 0.2$  and  $3.37 \pm 0.2$  Ga (Fig. 3). These ages overlap with those from metatuffs and ash beds of the upper Onverwacht Group

**Table 1.** Hf isotope data of the age populations of sample FT and MO

Age population ( $\pm 30$ Ma)	$\epsilon\text{Hf}_i$ (av.)	$\epsilon\text{Hf}_i$ (min.)	$\epsilon\text{Hf}_i$ (max.)	<i>n</i>
<i>Sample FT</i>				
3224	-1.3	-1.3	-	1
3299	-0.2	-5.9	+1.6	20
3365	+0.1	-1.8	+2.6	6
3465	+0.3	-2.0	+2.4	21
3534	+1.6	-0.5	+2.5	9
<i>Sample MO</i>				
3227	-1.8	-6.0	+0.3	34
3280	-0.9	-3.8	+1.1	37
3365	+0.5	0.0	+0.9	2
3459	+1.0	-1.7	+4.5	44
3519	+1.0	-3.3	+3.8	13
3650	+2.1	-	+2.1	1

(Kromberg Formation:  $3.334 \pm 0.006$  Ga; Mendon Formation:  $3.298 \pm 0.006$  Ga) and from a granitoid sample ( $3.323 \pm 0.007$  Ga) of the Ngwane gneiss of Swaziland (Byerly *et al.* 1996; Zeh *et al.* 2011). A similar age was also obtained from gneiss xenoliths in the Nelspruit 'granite' (Kamo & Davis 1994). These ages are not regionally specific: volcanic rocks of the Kromberg Formation occur only south of the Inyoka Fault, whereas Mendon volcanic rocks are exposed on either side of the Inyoka Fault, thus forming an overlap assemblage (Byerly *et al.* 1996). Thus, the 3.3 Ga zircons cannot serve well to distinguish a southern from a northern provenance.

The general absence of zircons with ages  $<3.265$  Ga in the Fig Tree greywacke of the Sheba Formation indicates that the latter does not contain material from reworked 3.259 Ga dacites, which is widespread in the lower Fig Tree Group south of the Inyoka Fault (e.g. Kröner *et al.* 1991; Byerly *et al.* 1996). Sheba Formation greywackes are thus either older than  $3.259 \pm 0.003$  Ga or had a different source. One or several restricted, well-defined source areas would be supported by the homogeneous age population of 3.45 Ga from a sample of southern-facies Fig Tree greywacke (Kröner & Compston 1988), whereas the northern facies sampled here is much more diverse. Considering the geological constraints and the obtained age spectra, we estimate the deposition age of Sheba greywacke at the sampled locality to be *c.*  $3260 \pm 10$  Ma. However, a younger deposition age cannot completely be excluded, as a single grain of sample FT yielded a younger U–Pb age of  $3.222 \pm 0.017$  Ga (grain a6). That single age, however, may reflect the timing of zircon alteration (see the irregular microstructure of grain a6 in Fig. 2a), accompanied by multiple (concordia-parallel) Pb loss. In any case, the obtained ages are in good agreement with the suggested depositional age of 3.359–3.225 Ga for the Fig Tree Group (Kröner *et al.* 1991; Byerly *et al.* 1996).

The Moodies Group sample contains, like sample FT, a major zircon age population at about  $3.28 \pm 0.02$  Ga (Fig. 3), but also three younger age groups: abundant zircons at  $3.228 \pm 0.008$  Ga, four grains at  $3.19 \pm 0.02$  Ga, and a single grain at  $3.095 \pm 0.024$  Ga. Apart from the youngest age, all other data are in good agreement with the suggested depositional age of 3.225–3.210 Ga for the Moodies Group (Heubeck *et al.* 2010). Sources of the youngest Moodies Group zircons (3.28–3.19 Ga) may include dacitic rocks of the lower Fig Tree Group south of the Inyoka Fault (Mapepe Formation:  $3.259 \pm 0.003$  Ga), and the upper Fig Tree Group on either side of the Inyoka Fault (Schoongezicht Formation:  $3.225 \pm 0.003$  Ga; Armstrong *et al.* 1990; Kröner *et al.* 1991; Kamo & Davis 1994; Byerly *et al.* 1996; de Ronde & Kamo 2000). Additional sources may have been the Kaap Valley and Nelshoogte Tonalites north of the Barberton Greenstone Belt ( $3227 \pm 1$  Ma; e.g. Kamo & Davis 1994), granitoids in the southwestern Barberton Greenstone Belt (Dalmein Pluton,  $3216 \pm 2$  Ma; Kamo & Davis 1994) or abundant granites of the Usutu Suite in Swaziland ( $3236$ – $3220$  Ma; Schoene *et al.* 2008; Zeh *et al.* 2009, 2011; Schoene & Bowring 2010). This wide range of potential sources and regions is importantly limited by involving the Hf isotope data. These point to a southern provenance, as is supported by the overlap of the Hf isotope data of the detrital zircons with those of the 3.54–3.23 Ga granitoids of the Barberton South Terrane and from Swaziland (Amelin *et al.* 2000; Zeh *et al.* 2009, 2011). In contrast, granitoids of the Barberton North Terrane are more radiogenic and plot on a different trend (Fig. 5).

In summary, the combined U–Pb and Hf isotope datasets clearly show that zircons of the Fig Tree Group greywacke and Moodies Group quartzarenite samples were originally supplied from similar basement sources to the south of the Inyoka Fault system. Because both samples contain zircon populations with nearly identical Hf isotope data and similar age populations (at 3.53, 3.47, 3.36 and

3.28 Ga), sediments of both units either had access to the same source area or Moodies provenance included Fig Tree Group greywackes, regardless of their location. The common presence of unambiguous and specific Fig Tree clasts in Moodies conglomerates, including banded ferruginous chert and banded iron formation, shows that a variety of Fig Tree Group lithologies were indeed reworked during Moodies Group deposition but that only those most resistant to chemical weathering escaped mechanical disaggregation during transport, tidal reworking and intensive chemical weathering (Hessler & Lowe 2006). In fact, reworking of Fig Tree Group sediments in Moodies time would also explain the seeming contradiction of southern provenance indicated by zircon U–Pb–Hf isotope data and partial northern sources indicated by sedimentological data.

### Archaean crustal evolution

The  $\epsilon\text{Hf}_t$  values of the detrital zircons overlap well with those of granitoids from the Barberton South Terrane and Swaziland (Fig. 5), and nearly all data plot in a gap between the crustal evolution trend of the oldest granitoids from the Ancient Gneiss Complex of Swaziland (AGC trend in Fig. 5) and the granitoids from the Barberton North Terrane (BNT trend). Figure 5 also shows that the 3.53 and 3.47 Ga zircon populations show wide  $\epsilon\text{Hf}_t$  variations between +4.5 and –1.7; that is, between values of depleted mantle and AGC crust. These values and variations indicate that new crust was formed from a depleted mantle source during both events, but also that crust–mantle mixing and crust reworking occurred in the hinterland at the same time. Hafnium model ages show that the reworked crust was on average not older than 3.75 Ga (see Fig. 5) except for one grain that yields a model age of 3.95 Ga (grain F52). The dataset also hints that new crust formation ceased after 3.45 Ga and that crustal reworking was the predominant process during 3.27 and 3.23 Ga magmatism (mostly subchondritic  $\epsilon\text{Hf}_t$ ), even though new crust formation between 3.45 and 3.23 Ga cannot completely be excluded. In fact, the observed  $\epsilon\text{Hf}_t$  variations could be explained either by reworking of a pre-existing heterogeneous crust or by the mixing of crust–mantle matter. Whatever the cause,  $\epsilon\text{Hf}_t$  values of the 3.23 Ga detrital zircons of sample MO are significantly lower than those of the 3.23 Ga TTGs of the Barberton North Terrane, which show superchondritic  $\epsilon\text{Hf}_t$  at the same time (Fig. 5). This is an additional indication that the detrital zircons of the Moodies sample were not derived from the Barberton North Terrane but rather from south of the Inyoka Fault. In fact, the new dataset supports previous conclusions that ancient, more radiogenic crust of the Barberton South Terrane was reworked during terrane amalgamation at about 3.25 Ga whereas the Barberton North Terrane was affected by some new crust formation and/or by the reworking of a younger, less radiogenic crust at the same time (Schoene *et al.* 2008, 2009; Zeh *et al.* 2009, 2011).

Detrital zircons of both samples document Palaeoarchaean crust–mantle evolution but show no evidence of juvenile Eoarchaean or even Hadean crust in the source area. Thus, evidence for such an old hinterland in southern Africa is at present documented only by detrital zircons from *c.* 3.1 Ga sediments of the Limpopo Belt (Fig. 1), of which some zircons show U–Pb ages up to 3.9 Ga and hafnium model ages up to 4.45 Ga (Zeh *et al.* 2008).

### Conclusions

Results of combined U–Pb and Lu–Hf isotope analyses on two samples show that Fig Tree Group greywacke and Moodies Group quartzarenite of the Barberton Greenstone Belt contain similar detrital zircon populations with ages at 3.53, 3.47, 3.36 and 3.28

Ga, and with overlapping Hf isotope compositions. The Moodies Group quartzarenite also contains a younger age population at 3.22–3.19 Ga. The source area of both sediments is characterized by new crust formation from depleted mantle sources at 3.53, 3.47, and perhaps at 3.36 Ga, accompanied by the partial reworking of older crust with Hf model ages between 3.95 and 3.75 Ga, and by significant crust reworking between 3.28 and 3.22 Ga. These data support the interpretation that zircons in both samples were originally dominantly derived from igneous rocks south of the Inyoka Fault, comprising the Stolzburg, Steynsdorp and Swaziland (sub) terranes. However, it remains unclear whether the detritus was continuously supplied from the same crustal source during deposition of both sediments or if Moodies Group quartzarenite was mainly derived from intensively reworked Fig Tree Group greywacke during extensive syn- and postorogenic strike-slip faulting and high uplift or subsidence between 3.26 and 3.19 Ga. In fact, reworking of Fig Tree sediments could solve the conundrum that some Moodies Group sediments reveal a southward directed transport, whereas detrital zircons require the opposite transport direction. A southern provenance for the Fig Tree and Moodies Group sediments also implies that Archaean basement rocks south of the Inyoka Fault were exposed at about 3.26 Ga, and that these basement rocks formed part of a morphological high (cordillera?) that perhaps existed until the end of terrane amalgamation at about 3.23 Ga. Thus, the results of this study demonstrate that combined U–Pb and Lu–Hf isotope data of detrital zircons not only set tight constraints on sediment provenances, but also can be useful for palaeo-landscape reconstruction at the time of sediment deposition.

## Appendix

### U–Th–Pb isotope analyses

Uranium, thorium and lead isotopes were analysed using a ThermoScientific Element 2 sector field (SF)-ICP-MS system coupled to a Resolution M-50 (Resonetics) 193 nm ArF excimer laser (ComPexPro 102F, Coherent) system at Goethe-University Frankfurt. Data were acquired in time-resolved peak-jumping pulse-counting/analogue mode over 356 mass scans, with a 20 s background measurement followed by 21 s sample ablation. Laser spot size was 23  $\mu\text{m}$  for unknowns and 33  $\mu\text{m}$  for the standard zircons GJ1, 91500, and Plešovice. The sample surface was cleaned directly before each analysis by four pulses pre-ablation. Ablations were performed in a 0.6 l  $\text{min}^{-1}$  He stream, which was mixed directly after the ablation cell with 0.07 l  $\text{min}^{-1}$   $\text{N}_2$  and 0.68 l  $\text{min}^{-1}$  Ar before introduction into the Ar plasma of the SF-ICP-MS system. All gases had a purity of >99.999% and no homogenizer 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  $^{254}\text{UO}/^{238}\text{U}$ , below 0.5%. The sensitivity achieved was in the range of 9000–14000 c.p.s.  $\mu\text{g g}^{-1}$  for  $^{238}\text{U}$  with a 23  $\mu\text{m}$  spot size, at 5.5 Hz and 5–6 J  $\text{cm}^{-2}$  laser energy. The typical penetration depth was about 14–20  $\mu\text{m}$ . The two-volume ablation cell (Laurin Technic, Australia) of the M50 allows detection and sequential sampling of heterogeneous grains (e.g. growth zones) during time-resolved data acquisition, owing to its quick response time of <1 s (time until maximum signal strength was achieved) and wash-out (<99.9% of previous signal) time of about 2 s. With a depth penetration of *c.* 0.7  $\mu\text{m s}^{-1}$  and a 0.46 s integration time (four mass scans = 0.46 s = one integration) any significant variation of Pb/Pb and U/Pb on the micrometre 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<sup>®</sup> spreadsheet program (Gerdes & Zeh 2006, 2009). A common-Pb correction based on the interference- and background-corrected  $^{204}\text{Pb}$  signal and a model Pb composition (Stacey & Kramers 1975) was carried out. The  $^{204}\text{Pb}$  content for each ratio was estimated by subtracting the average mass 204 signal, obtained during the 20 s baseline acquisition, which mostly results from  $^{204}\text{Hg}$  in the carrier gas (*c.* 180–420 c.p.s.), from the mass 204 signal of the respective ratio. For the analysed sample the calculated common  $^{206}\text{Pb}$  contents were mostly <1–2% of the total  $^{206}\text{Pb}$  but in rare cases exceeded 20%. 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 ( $^{206}\text{Pb}^*/^{238}\text{U}$ ) during the 21 s of sample ablation was corrected for each analysis. The correction was done by applying a linear regression through all measured, common Pb corrected ratios, excluding the outliers ( $\pm 2$  standard deviation; 2SD), and using the intercept with the *y*-axis as the initial ratio. The total offset of the measured drift-corrected  $^{206}\text{Pb}^*/^{238}\text{U}$  ratio from the ‘true’ isotope dilution thermal ionization mass spectrometry (ID-TIMS) value ( $0.0983 \pm 0.0004$ ; ID-TIMS GUF value) of the analysed GJ-1 grain was around 10% and the drift over the day was less than 2%.

Reported uncertainties ( $2\sigma$ ) of the  $^{206}\text{Pb}/^{238}\text{U}$  ratio were propagated by quadratic addition of the external reproducibility (2SD) obtained from the standard zircon GJ-1 ( $n = 21$ ; 2SD *c.* 1.3%) during the analytical session (*c.* 6 h) and the within-run precision of each analysis (2SE; standard error). For the  $^{207}\text{Pb}/^{206}\text{Pb}$  ratio we used a  $^{207}\text{Pb}$  signal-dependent uncertainty propagation (see Gerdes & Zeh 2009). The  $^{207}\text{Pb}/^{235}\text{U}$  ratio is derived from the normalized and error propagated  $^{207}\text{Pb}/^{206}\text{Pb}$  and  $^{206}\text{Pb}^*/^{238}\text{U}$  ratios assuming a  $^{238}\text{U}/^{235}\text{U}$  natural abundance ratio of 137.88 and the uncertainty derived by quadratic addition of the propagated uncertainties of both ratios. The accuracy of the method was verified by analyses of reference zircon 91500 ( $1066.4 \pm 5.8$  Ma, MSWD of concordance and equivalence = 0.28,  $n = 8$ ), and Plešovice ( $338.1 \pm 2.3$  Ma, MSWD<sub>C+E</sub> = 0.86,  $n = 7$ ). The data were plotted using the software Isoplot (Ludwig 2001).

### Lu–Hf isotope analyses

Hafnium isotope measurements were performed with a ThermoFinnigan NEPTUNE multicollector ICP-MS system at GUF coupled to the same laser as described in the U–Pb method (Resolution M-50 193 nm ArF excimer laser). Rectangular laser spots with edge lengths of 43  $\mu\text{m}$  were drilled with repetition rate of 5.5 Hz and an energy density of 6 J  $\text{cm}^{-2}$  during 55 s of data acquisition. All data were adjusted relative to the JMC475  $^{176}\text{Hf}/^{177}\text{Hf}$  ratio of 0.282160 and quoted uncertainties are quadratic additions of the within-run precision of each analysis and the reproducibility of JMC475 (2SD = 0.0033%,  $n = 8$ ). Accuracy and external reproducibility of the method were verified by repeated analyses of reference zircon GJ-1, MudTank and Temora2, which yielded  $^{176}\text{Hf}/^{177}\text{Hf}$  of  $0.282012 \pm 0.000024$  (2SD,  $n = 26$ ),  $0.282494 \pm 0.000026$  ( $n = 12$ ), and  $0.282660 \pm 0.000026$  ( $n = 7$ ), respectively. This is well within the range of solution mode data (Woodhead & Hergt 2005; Gerdes & Zeh 2006).

For calculation of the epsilon Hf ( $\epsilon\text{Hf}_t$ ) the chondritic uniform reservoir (CHUR) was used as recommended by Bouvier *et al.* (2008;  $^{176}\text{Lu}/^{177}\text{Hf} = 0.0336$  and  $^{176}\text{Hf}/^{177}\text{Hf} = 0.282785$ ), and a decay constant of  $1.867 \times 10^{-11}$  (average of Scherer *et al.* 2001; Söderlund *et al.* 2004). Initial  $^{176}\text{Hf}/^{177}\text{Hf}_i$  and  $\epsilon\text{Hf}_i$  for all analysed zircon domains were calculated using the apparent Pb–Pb ages obtained for the respective domains. Depleted mantle hafnium model ages ( $T_{\text{DM}}$ ) were calculated using values for the depleted

mantle as suggested by Blichert-Toft & Puchtel (2010), with  $^{176}\text{Hf}/^{177}\text{Hf} = 0.283294$  and  $^{176}\text{Lu}/^{177}\text{Hf} = 0.03933$ , corresponding to a straight depleted mantle (DM) evolution line with  $\epsilon\text{Hf}_{\text{today}} = +18$  and  $\epsilon\text{Hf}_{4.558\text{ Ga}} = 0.0$ .  $T_{\text{DM}}$  for all data were calculated by using the measured  $^{176}\text{Lu}/^{177}\text{Hf}$  of each spot for the time since zircon crystallization, and a mean  $^{176}\text{Lu}/^{177}\text{Hf}$  of 0.0113 for the Palaeoproterozoic–Archaean crust (mean of average continental crust as suggested by Taylor & McLennan (1985) and Wedepohl (1995)).

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