Detrital zircon in a supercontinental setting: Locally derived and far-transported components in the Ordovician Natal Group, South Africa

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Abstract: U-Pb and Lu-Hf signatures of detrital zircon from conglomerates and sandstones of the Ordovician Natal Group, South Africa were determined using LA-ICPMS. The basal conglomerates are dominated by Palaeo- to Mesoarchaean detrital zircon with ε_{Hf}-values from +3 to -4 with minor Mesoproterozoic input, indicating a proximal source in the Kaapvaal Craton and minor input from rocks of the Natal Sector of the Mesoproterozoic Namaqua-Natal province. The sandstones are all dominated by a combination of juvenile Mesoproterozoic zircon and Neoproterozoic zircon derived from Mesoproterozoic rocks that were reworked during the Pan–African Orogeny. Several sedimentary sequences from former Gondwana with Neoproterozoic to Permian depositional ages show similar detrital zircon signatures. Sedimentary sequences of such vast temporal and geographical distribution are unlikely to have been fed by a single source, making it more likely that these sequences were fed by several different (Pan–Gondwana) source terranes with closely similar U-Pb and Lu-Hf zircon signatures. The results show that source terrane non-uniqueness can make ascertaining sedimentary provenance from detrital zircon impossible, and should be taken as a reminder when using detrital zircon as evidence for far reaching conclusions in basin evolution studies and palaeogeography.

Supplementary material: U-Pb and Lu-Hf data are available at http://www.geolsoc.org.uk/SUP0000

Zircon is a highly refractory and robust mineral, and the U–Pb and Lu–Hf isotope systems of detrital zircon in a sediment or sedimentary rock are excellent recorders of the crystallisation age and evolution of the (proto)source rock, i.e. the igneous or metamorphic rock in which the zircon originally formed (e.g. Hawkesworth & Kemp 2006). These properties are thought to provide distinctive source terrane signatures (e.g. Williams 2001), and are therefore utilized in sedimentary provenance studies (e.g. Veevers *et al.* 2005, 2006; Augustsson *et al.* 2006; Veevers & Saeed 2007), whose ultimate goal is to trace the detrital zircon population of sedimentary rocks (commonly sandstone) back to their (mainly granitoid) sources (e.g. Fedo *et al.* 2003). Some studies have, however, indicated that the direct link between detrital zircon age and Hf isotope and source terrane may be questionable — because of recycling of older sediments (e.g. Andersen *et al.* 2011; Kristoffersen *et al.* 2014), which facilitates mixing of detritus from different sources, and because the U–Pb age and Hf isotope signature of various source terranes may not be sufficiently characteristic for them to be distinguishable (Andersen 2014).

With a long and well-preserved record of clastic sedimentary rocks ranging in age from Neoproterozoic to Recent, the older of which (Neoproterozoic to Triassic) formed in a Gondwana setting (Catuneanu *et al.* 2005; Shone & Booth 2005; Gresse *et al.* 2006), South Africa provides an ideal testing ground for detrital zircon as a provenance indicator. The importance of factors such as sedimentary recycling and indistinct source characteristics can be evaluated by detailed studies of well-preserved deposits, of which stratigraphy, depositional environment and tectonic setting are well constrained. Based on detrital zircon U-Pb and Lu-Hf data from Permian Vryheid Formation sandstones of the main Karoo basin in eastern South Africa, Veevers and Saeed (2007) suggested that detritus was transported from primary sources in central Antarctica to depositional basins in present-day southern Africa and adjacent parts of the Gondwana supercontinent in late Palaeozoic time.

The sandstones and conglomerates of the Ordovician Natal Group (Thomas *et al.* 1992; Marshall 2006) predate the Karoo sediments studied by Veevers and Saeed (2007) by ca. 200 Ma. These rocks thus hold a potential key role for the understanding of the detrital zircon budget of Phanerozoic sedimentary sequences in SW Gondwana: If significant amounts of material from central Antarctica was transported to southern Africa in Permian time, detrital zircon in the older Natal Group should show age and Hf isotope characteristics significantly different from that of the Vryheid Formation sandstones. Furthermore, the depositional basin of the Natal Group was located close to possible sources in Antarctica and Mozambique, and it was deposited on a variety of basement rocks, including Archaean rocks of the Kaapvaal craton and various igneous and metamorphic rocks of the late Mesoproterozoic Natal Sector of the Namaqua-Natal Province. Contributions from local basement sources would therefore show distinctive detrital zircon signatures.

In the present study, we have analysed detritial zircon from selected sedimentary rocks of the Natal Group to provide an independent test of the provenance model of Veevers and Saeed (2007), and to evaluate alternative sources of detritus in southern Africa and adjacent parts of Gondwana. Since the Mesoproterozoic basement rocks of eastern South Africa are very poorly constrained by published Hf isotope data, it has been necessary also to perform a pilot study on granitoid plutons that make up a substantial and zircon-fertile component of the regional basement.

Geological setting

The geology of southern Africa is dominated by the Archaean Kaapvaal Craton which preserves a record of c. 1000 Ma of geological history spanning from c. 3600 to c. 2700 (e.g. Hunter *et al.* 2006). It comprises largely granitoid rocks with a tonalite-trondhjemite-granodioritic composition (Hunter *et al.* 2006; Robb *et al.* 2006), infolded greenstone belts and their remnants, which include the well-studied Barberton Greenstone Belt (Brandl *et al.* 2006), and volcano-sedimentary cover sequences (Hunter *et al.* 2006) such as the Pongola

Supergroup (e.g. Gold 2006), the Dominion Group (e.g. Marsh 2006), and the Witwatersrand (e.g. McCarthy 2006) and Ventersdorp Supergroups (e.g. van der Westhuizen *et al.* 2006). To the north the Kaapvaal Craton borders the Neoarchaean Limpopo Belt, which comprises mostly gneisses (Kramers *et al.* 2006). The granitoid rocks and greenstone belts of the Kaapvaal Craton, as well as some sedimentary rocks, and the Limpopo Belt are well covered by both U-Pb and Lu-Hf data (Zeh *et al.* 2007, 2008, 2009, 2010, 2011, 2012, 2013; Gerdes & Zeh 2009), while for the volcano-sedimentary cover sequences Lu-Hf data are only available for the Eldorado Reef of the uppermost Witwatersrand Supergroup (Koglin *et al.* 2010). At its southern and western margin the Kaapvaal Craton is bounded by the Namaqua-Natal Province (e.g. Cornell *et al.* 2006).

The Namaqua-Natal Province is a Mesoproterozoic orogenic belt related to the amalgamation of the supercontinent Rodinia (e.g. Cornell *et al.* 2006). It forms a continuous belt from the Namaqua Sector in southern Namibia and NW South Africa (Northern Cape province) to the Natal Sector in eastern South Africa (KwaZulu-Natal province), with the between-lying area covered by the Carboniferous to Triassic Karoo Supergroup (Cornell *et al.* 2006). Its southern boundary is formed by the Neoproterozoic to early Cambrian Saldania Belt (e.g. Cornell *et al.* 2006). Sm-Nd work has shown that, while the Natal Sector has a juvenile nature (Eglington *et al.* 1989; Thomas & Eglington 1990), the Namaqua Sector has a more prolonged crustal history (Yuhara *et al.* 2001; Pettersson *et al.* 2009). The juvenile nature of the Natal sector is also supported by Hf data (Spencer *et al.* 2015). The southern two-thirds of the Natal Sector are dominated by granitoid rocks which make up >80% of the exposed area (Eglington *et al.* 2003). The Oribi Gorge Suite, which consists of 10 rapakivi–charnockite plutons emplaced in two episodes of intrusion at ~1070 and ~1030 Ma, is the most extensive of these granitoids (Eglington *et al.* 2003).

From the end of the Pan-African orogeny in late Neoproterozoic time southern Africa remained within a stable supercontinental setting, Gondwana, until its breakup in Jurassic time (e.g. Torsvik & Cocks 2013). Neighbours during this time were South America, the Falkland Plateau, the Ellsworth-Whitmore Mountains and Dronning Maud Land (Torsvik & Cocks 2013). In this setting continental cover sequences were deposited, ranging in depositional age from Neoproterozoic to Triassic. In the Early Ordovician Period (480 Ma) South Africa was located at ~20° S, and, as a result of southward drift, it was located at ~35° S (Torsvik & Cocks 2013) by the Late Ordovician Period (445 Ma). Prior to this, in the Neoproterozoic to Cambrian Periods, the western part of South Africa was affected by the Saldanian orogeny, leading to granite emplacement (e.g. Scheepers & Armstrong 2002; Chemale Jr. *et al.* 2010; Villaros *et al.* 2012), deposition of sedimentary rocks (e.g. Gresse *et al.* 2006) and the development of the Cape basin (Tankard *et al.* 2009). Deposition in the Cape basin lasted from the Ordovician to the Early Carboniferous Period (Tankard *et al.* 2009); the Ordovician Natal Group is contemporaneous with at least some of this (Cape Supergroup) succession (Shone & Booth 2005; Tankard *et al.* 2009).

Well-preserved cover sequences cropping out in South Africa for which U-Pb (and some Lu-Hf) data are available include the Neoproterozoic Cango Caves and Kansa Groups (Naidoo *et al.* 2013), the Neoproterozoic Nama Group (Blanco *et al.* 2011), Neoproterozoic sedimentary rocks from the Saldania Belt (Frimmel *et al.* 2013), the early Palaeozoic Cape Supergroup (Fourie *et al.* 2010; Vorster 2013), and the Carboniferous-Triassic Karoo Supergroup (Veevers & Saeed 2007; Vorster 2013). Veevers & Saeed (2007) argued for a distant (central Antarctic) source for late Mesoproterozoic and Neoproterozoic zircon in Permian sandstones from the main Karoo basin in eastern South Africa (province of KwaZulu-Natal). A distant provenance (Mozambique) has also been suggested for the Ordovician Natal Group based on palaeocurrent data indicating a source to the north and NE of the depositional basin (Thomas *et al.* 1992; Marshall 2002) and a c. 580 Ma K–Ar age recorded for detrital muscovite (Thomas *et al.* 1992).

The Natal Group

The mainly maroon-coloured arkosic sandstones and quartz arenites, with interbedded mudrock and conglomerate units, of the Ordovician Natal Group were deposited on Archaean to Mesoproterozoic basement of the Kaapvaal Craton and the Natal Sector of the Namaqua-Natal Province (Hobday & von Brunn 1979; Marshall 2002). The geometry and tectonic setting of the Natal Group sedimentary basin is uncertain, having been referred to as a half-graben foreland basin (Hobday & von Brunn 1979), a foreland graben basin (Marshall 2002), a divergent margin basin (Shone & Booth 2005), and a transpressional foreland basin (Tankard *et al.* 2009). The main outcrops of the Natal Group are found within the province of KwaZulu-Natal (South Africa), from Hlabisa (Fig. 1) in the north to just south of Hibberdene (Marshall 2006) in the south. The southern limit of the Natal Group basin has traditionally been defined by the Dweshula High (Fig. 1; e.g. Marshall 2002, 2006), smaller outcrops of the Natal Group occuring as much as 30 km south of this palaeo-topographic high are, however, also known (Marshall 2002; Hicks 2010). North of the Tugela Thrust Front (Fig. 1) the Natal Group rests on Archaean rocks of the Kaapvaal Craton, whereas to the south it rests nonconformably on the Namaquan (Mesoproterozoic) granitoids and gneisses of the Natal Sector (Marshall 2006). It is generally disconformably overlain by the Carboniferous-Permian Dwyka Group of the Karoo Supergroup (Marshall 2006), except for south of the Dweshula High (Fig. 1) where it is occasionally unconformably overlain by the Devonian Msikaba Formation (Hicks 2010).

The Natal Group (Fig. 2) comprises the Durban Formation, which is subdivided into the Ulundi, Eshowe, Kranskloof, Situndu, Dassenhoek and Melmoth Members, and the Mariannhill Formation, which is subdivided into the Tulini, Newspaper and Westville Members (Marshall 2002, 2006). The Natal Group sediments are interpreted to have been deposited in at least two cycles of sedimentation, the first being initiated with the deposition of the conglomerates and interbedded sandstones of the Ulundi Member and closed by the initiation of the second cycle with deposition of the small-pebble conglomerates of the Tulini Member (Marshall 2006). A possible third cycle of sedimentation was initiated with the deposition of the Westville Member, the product of which might have been, to a large extent, removed by erosion prior to the deposition of the glaciogenic sediments of the Permian Dwyka Group (Karoo Supergroup) (Marshall 2006). Marshall (2006) interpreted these cycles to have been caused by uplift of the provenance area, which he thought to have been located in the Pan-African belt of southern Mozambique (Thomas *et al.* 1992; Marshall 2002, 2006), or by renewed subsidence of the depositional basin. While a mainly fluvial origin of the Natal Group (Hobday & von Brunn 1979; Marshall & von Brunn 1999; Marshall 2002, 2006) is generally accepted, the origin of the silicified quartz arenites of the Kranskloof and Dassenhoek Members is somewhat disputed (e.g. Liu & Cooper 1998; Marshall 1999). These two members have been interpreted as fluvial and eolian reworking of the fluvial Eshowe Member (Marshall & von Brunn 1999; Marshall 2002; 2006), and as shallow-marine, tidally-influenced sequences (Liu & Cooper 1998).

Material studied

Eight sedimentary rock samples were analysed, comprising one quartzite clast and one matrix-dominated sample from the basal Ulundi Member conglomerate, and sandstone samples from the Ulundi, Eshowe, Dassenhoek, Tulini and Newspaper Members. These samples were chosen to provide a reasonable coverage of both geographic location and stratigraphic level. Sample localities are given in Figure 1, and their coordinates are listed in Table 1. U–Pb data of the sedimentary rocks are given in Figure 3, and Lu–Hf data are given in Figure 4 and 5.

In order to provide some Hf data for comparison with potential local bedrock sources, two granite samples — SA12/24 and SA13/133, belonging to the Mgeni and Fafa plutons, respectively, of the Oribi Gorge Suite within the Mzumbe Terrane of the Natal Sector — were dated (Fig. 6) and their Lu–Hf content was analysed (Fig. 4). This data, combined with the Hf data of Spencer *et al.* (2015) provides an extensive Hf signature of the zircon-fertile lithologies in the region.

Analytical methods

The samples were crushed, and their heavy mineral fractions were extracted by pan-washing, and, in the cases of SA12/06 and SA12/28, heavy liquid (sodium heteropolytungstate) separation. No magnetic separation was performed to avoid introducing an artificial bias (Sircombe & Stern 2002; Andersen *et al.* 2011). Random selections of zircon grains were picked, cast in epoxy resin, polished and imaged by cathodoluminescence (CL) using a JEOL JSM 6460LV scanning electron microscope at the Department of Geosciences, University of Oslo.

U–Pb and Lu–Hf analyses were done by LA-ICPMS, using a Nu Plasma HR multi-collector mass spectrometer equipped with a NewWave LUV 213 Nd-YAG and a Cetac LSX-213 G2+ laser microprobe at the Department of Geosciences, University of Oslo. The analytical protocols of Andersen *et al.* (2009) and Rosa *et al.* (2009) were followed for U–Pb, whereas for Lu–Hf those of Elburg *et al.* (2013) were used. Ablation conditions for the NewWave LUV 213 laser microprobe were: beam diameter 40 μm (aperture imaging mode), pulse frequency 10 Hz and beam fluence c. 0.06 J/cm^2 for U–Pb, and beam diameter 55-60 μm (aperture imaging mode), pulse frequency 5 Hz and beam fluence c. 2 J/cm^2 for Lu–Hf; in both cases using static ablation. For the Cetac LSX-213 G2+ laser microprobe ablation conditions were: beam diameter 40 μm (aperture imaging mode), pulse frequency 10 Hz and beam fluence < 0.78 J/cm^2 for U–Pb, and beam diameter 50 μm (aperture imaging mode), pulse frequency 5 Hz and beam fluence c. 15 J/cm^2 for Lu–Hf; in both cases using static ablation. Data reduction was done using an interactive, in-house Microsoft Excel 2003 spreadsheet programme for U–Pb, and using Nu Instruments built-in software for Lu–Hf.

U-Pb ages were calculated using the decay constants of Steiger & Jäger (1977). Discordance percentage were calculated as described in Kristoffersen *et al.* (2014). Ages are given as ^{206}Pb–^{238}U ages if younger than or equal to 600 Ma, otherwise the ^{207}Pb–^{206}Pb ages are used. Only grains with less than ±10% central discordance are included.

Zircons GJ-1 (^{207}Pb–^{206}Pb age = 609±1 Ma; Jackson *et al.* 2004), 91500 (^{207}Pb–^{206}Pb age = 1065±1 Ma; Wiedenbeck *et al.* 1995) and A382 (concordia age = 1876±2 Ma; Huhma *et al.* 2012) were used as U–Pb standards. Repeated analyses of the in-house reference zircon C (ID-TIMS, weighted average ^{207}Pb–^{206}Pb age = 556.4 ± 1.5 Ma; J. Lamminen, pers. comm. 2011) during the period the samples were analysed gave a weighted average ^{207}Pb–^{206}Pb age of 559 ± 3 Ma (2σ, MSWD = 0.76, probability of fit =0.87, n = 44). During the period the samples were analysed, repeated analyses of the Mud Tank zircon ($^{176}Hf/^{177}Hf$=0.282507±6, solution analysis; Woodhead & Hergt 2005) yielded $^{176}Hf/^{177}Hf$=0.282509±30 (2SD; n=139) and the in-house reference zircon LV-11 ($^{176}Hf/^{177}Hf$=0.282830±28, solution analysis; Heinonen *et al.* 2010) yielded $^{176}Hf/^{177}Hf$=0.282827±55 (2SD; n=99), the latter (±2 ε_{Hf}) is accepted as a conservative estimate of the precision of the method. A decay constant value for ^{176}Lu of 1.867 × 10^{-11} year^{-1} (Söderlund *et al.* 2004), and the present-day chondritic $^{176}Hf/^{177}Hf$=0.282785 and $^{176}Lu/^{177}Hf$=0.0336 (Bouvier et al. 2008) have been used in all ε_{Hf} calculations. We have adopted the depleted mantle parameters of Griffin *et al.* (2000); this model, modified to the decay constant and CHUR parameters used, gives present-day $^{176}Hf/^{177}Hf$=0.28325 (+16.4 ε_{Hf}, similar to average mid-ocean ridge basalt) from a chondritic initial $^{176}Hf/^{177}Hf$ at 4.56 Ga and $^{176}Lu/^{177}Hf$=0.0388.

Data handling

Concordia plots and ages, and weighted average ages were calculated using Isoplot (Ludwig 2008). Histogram, kernel density estimate (KDE), empirical cumulative distribution function (ECDF) and ε_{Hf} plots were made using the R programming language and statistical computing environment (R Development Core Team 2015) with the *ggplot2* plotting system (Wickham 2009). 1D KDE functions were calculated using the algorithm of Botev *et al.* (2010); 1D gaussian KDE functions with a fixed bandwidth=30 were also produced. 2D KDE functions were calculated using the R-function *kde2d* from the MASS package (Venables & Ripley 2002), over an equally spaced 100x100 grid spanning from 400 to 4000 in the x-direction and -15 to 15 in the y-direction using fixed bandwidths of 30 and 1.5 in the x- and y-directions, respectively. 1D two sample Kolmogorov-Smirnoff (K-S) tests were calculated using the *ks.test* function in R. 2D two sample K-S tests were calculated using the MATLAB-function *kstest_2s_2d* (available from MATLAB Central). The two sample K-S test is a measure of the maximum vertical distance (D_{max}) between two ECDFs. A probability value (p) is calculated for D_{max} to determine if D_{max} is the result of random sampling error or a true difference in the data. If $p > \alpha$ (α typically 0.05 in detrital zircon provenance studies) it is deemed unlikely that the samples are from different populations, the reverse is true for $p < \alpha$.

KDE curves represent probability densities as a function of zircon age, and the probability of finding a zircon within any age interval is given by the corresponding area under the curve, i.e. integral of the KDE function *a(t)* over the age interval of interest (t_{min} to t_{max}): $\int_{t_{\min}}^{t_{\max}} a(t)dt$ in which *a(t)* is the KDE function of interest normalized to unit area (i.e. $\int_{-\infty}^{+\infty} a(t)dt = 1$). This integral must be solved numerically.

Satkoski *et al.* (2013) introduced a "likeness" parameter for pairs of detrital zircon age distributions defined by

$$L = 1 - \frac{1}{2}\sum_{i=1}^{N}|a_i - b_i| \tag{1}$$

where a_i and b_i are values of two KDE functions whose values are known at N points along the time axis. The summation term in equation (1) is a simple, stepwise numeric approximation to the integral of the difference between two KDE functions over the time interval of interest, i.e.:

$$\sum_{i=1}^{N}|a_i - b_i| \approx \frac{1}{\Delta t}\int_{t_{\min}}^{t_{\max}}|a(t) - b(t)|dt \tag{2}$$

where Δt is the distance between the adjacent, equally spaced points where the values of *a* and *b* in equation (1) were determined.

Bivariate U–Pb and Lu–Hf data have mainly been visualized in simple scatter plots or by plots of model ages derived from the data (e.g. Veevers & Saeed 2007), although contoured 3D probability density surfaces based on the KDE model have also been sporadically used (Andersen 2002, 2013). However, the likeness parameter of Satkoski *et al.* (2013) may easily be extended to bivariate data, summing over differences between pairs of values on 3D probability density surfaces over the age–ε_{Hf} plane taken at regularly spaced points in an M by N grid:

$$L_2 = 1 - \frac{1}{2}\sum_{j=1}^{M}\sum_{i=1}^{N}\left|a_{i,j} - b_{i,j}\right| \tag{3}$$

in which both probability density functions must be normalized to unit volume, i.e.

$$\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} a(t,\varepsilon_{Hf})\,dt\,d\varepsilon_{Hf} = 1\,.$$

As the likeness parameter of Satkoski *et al.* (2013) is a much more robust measure of similarity/dissimilarity between pairs of detrital zircon samples (Satkoski *et al.* 2013) the results of the likeness tests are preferred over those of K-S tests.

Results

Granites

Zircons in the Mgeni pluton sample (SA12/24) are discordant (Fig. 6a), but the 10 least discordant grains (-4% < discordance < 1%) give a weighted average ^{207}Pb–^{206}Pb age of 1061±8 Ma (95% confidence, MSWD=0.70); the sample has an ε_{Hf}-value of 3.7±1.1 (2σ) (Fig. 4). The Fafa pluton sample (SA13/133) gives a concordia age (Fig. 6b) of 1086±6 Ma (2σ, MSWD=0.00051) and has an ε_{Hf}-value of 7.2±1.7 (2σ) (Fig. 4). The positive ε_{Hf}-values for both of these intrusions agree with the generally juvenile nature of the Mesoproterozoic rocks in the Natal Sector of the Namaqua-Natal Province (Fig. 7) (e.g. Eglington *et al.* 1989; Thomas & Eglington 1990; Spencer *et al.* 2015).

Sedimentary rocks

The two conglomerate samples from the basal Ulundi Member are dominated by Palaeo- to Mesoarchaean (3600-2940 Ma) detrital zircon (Fig. 3), whose ε_{Hf}-values range from +3 to -4 (Fig. 5). In contrast, the six sandstone samples from the Ulundi, Eshowe, Dassenhoek, Tulini and Newspaper Members are all dominated by a late Mesoproterozoic to earliest Neoproterozoic (1150-950 Ma) age zircon (Fig. 3). An additional, smaller, Neoproterozoic (700-500 Ma) fraction of variable size is also found in all of the sandstone samples. The late Mesoproterozoic to earliest Neoproterozoic fraction has an overall ε_{Hf}-range of +11 to -13 (Fig. 4), with 95% (246/258) of the grains plotting above the CHUR (chondritic uniform reservoir) curve. The Neoproterozoic fraction has a more crustally influenced ε_{Hf}-signature, where 98% (148/151) of the zircons have ε_{Hf} between +6 and -6. Minor Archaean, Palaeoproterozoic and Mesoproterozoic fractions are also present in several of the sandstone samples. Pre-1400 Ma zircons are most abundant in the northern part of the outcrop area (samples SA12/39, SA12/27, SA12/42, SA12/43), while zircons of this age are almost absent from the southern samples (SA12/20, SA12/06).

Discussion

The detrital zircon age and Hf isotope distribution patterns in Figures 3 through 5 suggest that three main detrital zircon components are present in the rocks of the Natal Group. (1): Palaeo- to Meosarchaean, (2) Mesoproterozoic to earliest Neoproterozoic, and (3) Neoproterozoic age, with characteristic ε_{Hf}-ranges of +3 to -8, +11 to -13 and +10 to -11, respectively. To understand the overall provenance characteristics of the Natal Group, the origin of each of these components needs to be identified, and the differences in relative abundance between samples should be characterized.

Similarity and differences between samples

The most obvious inter-sample difference observed is between the Ulundi conglomerate samples with a high proportion Archaean grains on one hand, and all of the sandstone samples dominated by Proterozoic zircon on the other (Figs. 3, 4, 5). To quantify these relationships, univariate (age) and bivariate (age + Hf isotope) likeness values have been calcuated.

Pairwise one-dimensional (Table 2) and two-dimensional likeness values (Table 3) show a high degree of similarity among the sandstone samples, close to the average likeness expected from samples drawn from a single population (0.72±0.06 for n≈100; Satkoski *et al.* 2013). The likeness values between the two samples of Ulundi conglomerate (1D: 0.79, 2D: 0.81) suggest that the detrital zircon patterns of these samples are also indistinguishable. Likeness values comparing the conglomerate and sandstone samples (1D: < 0.14, 2D: < 0.13) confirm the significant difference between these groups suggested from visual inspection.

Compared to the results of the K-S tests (Table 4 and 5) and the ECDF plot (Figure 8) these results show that the absence of pre-1400 Ma zircon from the southern part of the outcrop area of the Natal Group may reflect the existence of Meso-Palaeoproterozoic sources to the north of the basin, which have not contributed in the south. On the other hand, if sedimentation in the north and south was not synchronous, sources of pre-1400 Ma zircon in the northern, earlier deposits could have been eroded away or blanketed over by the time the later, southern deposits were formed.

Potential source areas

The Archaean population of the two conglomerate samples (Fig 5) shows a clear similarity in its ε_{Hf}-values with values recorded in the Kaapvaal Craton (Zeh *et al.* 2009, 2011, 2013). Although the Limpopo Belt and the Zimbabwe Craton also have broadly similar signatures (Zeh *et al.* 2007, 2009, 2010; Gerdes & Zeh 2009) and can therefore not be ruled out by zircon data, the Kaapvaal Craton is the Archaean domain in closest proximity to the depositional basin of the Natal Group and is, consequently, the most likely source area for the Archaean detrital zircons recorded in the basal Ulundi conglomerate samples. The quartzite clast from the Ulundi conglomerate (SA12/28) only contains Archaean zircons and could thus be derived from the quartzites of the Mesoarchaean Pongola Supergroup (e.g. Wronkiewicz & Condie 1989; Gold 2006) on the Kaapvaal Craton. The Archaean detrital zircon component disappears up stratigraphy from the basal Ulundi conglomerate to the overlying sandstone. The reason could either be a change in input to the basin, or that Archaean source rocks of the Kaapvaal Craton were blanketed over by deposition of the first sediments of the Ulundi Member. In addition to its dominant Archaean population the Ulundi matrix sample (SA12/29) also contain three, near-concordant, middle Mesoproterozoic to earliest Neoproterozoic zircon grains which could be derived from the Natal Sector of the Namaqua-Natal Province where similar ages are recorded (e.g. Eglington 2006). This suggests that the source area shedding sediments to the Ulundi conglomerate was altogether proximal to the depositional basin.

The Archaean grains recorded in the sandstone samples have similar ε_{Hf}–values as those of the conglomerate samples (Fig. 4-5), varying from slightly subchondritic to slightly superchondritic. One exception is the small cluster of c. 2730-2710 Ma zircons with an ε_{Hf}-range of -7 to -8 (Fig. 4), which could point to protosource-rocks whose protoliths had an extended crustal history, such as seen in the Ancient Gneiss Complex of Swaziland (Zeh *et al.* 2011).

The largest age group recorded in this study, dominating all sandstone samples, is found at c. 1150-950 Ma and shows a consistent peak at c. 1030 Ma (Fig. 3). This time period corresponds with the Rodinia-related Namaquan Orogeny (e.g. Cornell *et al.* 2006). Rocks of this age are now exposed in the Natal and Namaqua Sectors of the Namaqua-Natal Province which stretches from the KwaZulu-Natal area in South Africa to southern Nambia. Sm–Nd data indicate that the (western) Namaqua Sector generally has a longer crustal history of the two domains (e.g. Yuhara *et al.* 2001; Pettersson *et al.* 2009); the only published ε_{Hf}-values from the (western) Namaqua Sector (Cornell *et al.* 2013) are consistent with these observations. The (eastern) Natal Sector has a much more juvenile signature (e.g. Eglington *et al.* 1989; Thomas & Eglington 1990). The superchondritic ε_{Hf}-signatures of the granites reported in this study, also found in the data of Spencer et al. (2015), add supporting evidence to the juvenile nature of the Natal Sector. The Namaqua Sector is therefore an unlikely source area for the generally juvenile, late Mesoproterozoic to earliest Neoproterozoic zircons found in the Natal Group (Fig. 4); granitoid rocks of the Natal Sector (e.g. McCourt *et al.* 2006) are however, possible sources. Similar ages are, for instance, prevalent in the intrusive rocks of the Oribi Gorge Suite (Eglington *et al.* 2003; this study). Juvenile rocks of this age are also found in the Mozambique belt in southern (Grantham *et al.* 2003) and northeastern (Bingen *et al.* 2007) Mozambique, in the Ellsworth-Whitmore Mountains, Antarctica (Flowerdew *et al.*

2007), in Dronning Maud Land, Antarctica (Bauer *et al.* 2003; Ramo *et al.* 2009) and on the Falkland Islands (Jacobs *et al.* 1999; Thomas *et al.* 2000). These areas were all adjacent to the depositional basin of the Natal Group (e.g. Torsvik & Cocks 2013) at its presumed depositional age of 490 Ma (Thomas *et al.* 1992).

Although the slightly smaller zircon population at c. 700-500 Ma (Fig. 3) is fairly consistent throughout the sandstone samples, there seems to be a tendency of increasing relative fractions of younger grains from north to south (Fig. 3). The bulk of the late Neoproterozoic to early Cambrian zircons have ε_{Hf}-values indicative of a derivation from juvenile, Mesoproterozoic material reworked during the Pan-African Orogeny, while the youngest grains recorded (< 550 Ma) could point to a source in the Cape Granite Suite, towards the SW within present-day South Africa (Villaros *et al.* 2012).

Implications

Veevers & Saeed (2007) observed detrital zircon patterns closely similar to those reported in this study in Permian sedimentary rocks in the main Karoo Basin of South Africa (Vryheid Formation) and in Dronning Maud Land, Antarctica. Their interpretation was that these sediments were derived from a common source in central Antarctica, which shed detritus to surrounding parts of Gondwana in the late Palaeozoic. And they inferred from Cambrian (meta)sediments with similar zircon signatures from the Welch Mountains, Antarctica and the Ellsworth-Whitmore Mountains that the same central Antarctica source was also active in the Cambrian period. However, apart from the Karoo, Dronning Maud Land, Welch Mountains, and the Ellsworth-Whitmore Mountains, similar detrital zircon signatures have been reported from sedimentary rocks in many parts of former Gondwana and over a range of depositional ages from Neoproterozoic to Permian, including southern Africa (Fourie *et al.* 2010; Blanco *et al.* 2011; Frimmel *et al.* 2013; Naidoo *et al.* 2013), Antarctica (Flowerdew *et al.* 2007; Vorster 2013), South America (Blanco 2010; Uriz *et al.* 2010; Ramos *et al.* 2014) and Australia (Veevers *et al.* 2006; Veevers & Saeed 2007). Several of these sedimentary sequences show a high degree of Satkoski-likeness to the Natal Group sandstones and amongst each other (Table 6 and 7). Detrital zircon with similar age and Hf isotopic characteristics must therefore have been present over large surface areas in Gondwana much earlier than the Permian. This suggests either (1): that there has been a source feeding this vast area at least from Neoproterozoic to Permian time, (2): that these deposits are the result of recycling of older sedimentary sequences (the signatures thereby being a manifestation of the inherent resilience of zircon to physical and chemical weathering) or (3): that the various areas of Gondwana that fed these sandstone sequences are not sufficiently distinctive for them to be distinguished from age and Hf isotope data. If the first alternative is true, a source, possibly the central Antarctica source of Veevers & Saeed (2007), must have fed sediments to Gondwana, at least semi-continuously, from its inception almost until its subsequent breakup. This would require an enormous amount of available material. As juvenile Mesoproterozoic rocks and juvenile Mesoproterozoic rocks reworked during Neoproterozoic time are a common Pan-Gondwanan feature (Fig. 9) (e.g. Beckinsale *et al.* 1977; Wareham *et al.* 2001; Jacobs *et al.* 2003; Viola *et al.* 2006; Ramo *et al.* 2009) the third, Pan-Gondwanan alternative is seen as the most likely, with a possible minor influence of the second, recycling alternative in, at least, some of these sequences.

Combined with the results of sedimentary sequences with similar detrital zircon patterns our results show that sedimentary sequences dominated by juvenile Mesoproterozoic zircon and Neoproterozoic zircon derived from juvenile, Mesoproterozoic material reworked during the

Pan-African Orogeny are a common feature of neighbouring areas of former southern Gondwana. Thus the Permian Karoo and Dronning Maud Land sandstones of Veevers & Saeed (2007) need not be derived from Cental Antarctica — similar signatures were abundant in adjacent areas from at least the Neoproterozoic — but could be the result of recycling, or could be derived from one of several (equally) likely source terranes.

Conclusion

Taken together these results (Fig. 3-5, 8; Table 2-5) show that the basal Ulundi conglomerates of the Natal Group are derived from the same proximal source, i.e. from late Archaean rocks of the Kaapvaal craton – possibly from the Pongola Supergroup. On the other hand, the stratigraphically higher sandstones in the sequence show uniform provenance characteristics, reflecting source areas with a typical Gondwanan signature comprising juvenile Mesoproterozoic material, and Neoproterozoic material derived from juvenile Mesoproterozoic protoliths reworked during the Pan-African Orogeny (Ireland *et al.* 1998; Goodge *et al.* 2002; Flowerdew *et al.* 2007; Veevers & Saeed 2007; Fourie *et al.* 2010; Blanco *et al.* 2011; Villaros *et al.* 2012; Naidoo *et al.* 2013). Rocks with such age and Hf isotope characteristics are widespread across Gondwana, and thus the detrital zircons cannot be assigned to a specific source terrane, be it in central Antarctica or elsewhere.

The results should be taken as a reminder of the difficulty of ascertaining a provenance for detrital zircon, caused by sedimentary recycling or source terrane non-uniqueness: special care must be taken when using detrital zircon as evidence for far reaching conclusions, such as basin evolution and palaeogeography.

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Figure captions

Fig. 1.

Map of the study area (after Marshall & von Brunn 1999; Marshall 2002) showing outcrops of the Natal Group and the Msikaba Formation with sample localities indicated.

Fig. 2.

Generalized stratigraphy of the Natal Group (Marshall & von Brunn 1999; Marshall 2002). Not drawn to scale.

Fig. 3.

Combined histogram and kernel density estimate plots of all samples stacked according to their (presumed) stratigraphic positions. Ages are given as ^{206}Pb–^{238}U ages if equal to or younger than 600 Ma, otherwise the ^{207}Pb–^{206}Pb ages have been used. Note that the individual panels use different y–axis scaling. Solid curves are KDEs calculated using the algorithm of Botev *et al.* (2010); dashed curves are gaussian KDEs with bandwidth=30.

Fig. 4.

Initial ε_{Hf} plotted against age for sandstone samples. Contours of a KDE surface for sample SA12/06 drawn at the 95th percentile, and initial ε_{Hf} vs. age for granite samples (squares: SA12/24, Mgeni pluton; diamonds: SA13/133, Fafa pluton) are added to all panels. CHUR: chondritic uniform reservoir; DM: depleted mantle.

Fig. 5.

Initial ε_{Hf} plotted against age for the Archaean zircon fraction of the conglomerate samples. Contours of KDE surfaces for comparison data of Archaean age from southern Africa drawn at the 95th percentile are added to each panel; vertical lines: Eldorado Reef, uppermost Central Rand Group (Koglin *et al.* 2010); horizontal lines: Limpopo Belt (Zeh *et al.* 2007, 2008, 2009, 2010; Gerdes & Zeh 2009); dark grey: Murchinson Northern Kaapvaal (Zeh *et al.* 2009, 2012, 2013); light grey: Barberton (Zeh *et al.* 2009). CHUR: chondritic uniform reservoir; DM: depleted mantle. 2D KDEs were calculated over an equally spaced 100x100 grid spanning from 2800 to 3800 in the x-direction and -5 to 5 in the y-direction using fixed bandwidths of 30 and 1.5 in the x- and y-directions, respectively.

Fig. 6.

Concordia plot of granite samples. Error ellipses are plotted as 2σ. (**a**) SA12/24. (**b**) SA13/133.

Fig. 7.

Histogram of individual zircon initial ε_{Hf} (this study; Spencer *et al.* 2015) and whole rock ε_{Nd} from basement rocks in the Natal Province. Black: $2\varepsilon_{Nd} + 2$; diagonal lines: ε_{Hf} data from Spencer *et al.* (2015); grey: ε_{Hf} data this study. ε_{Nd} data obtained from the DateView database (Eglington 2004). ε_{Nd} data: Eglington (Unpublished); Thomas & Eglington (1990); (Thomas *et al.* 1993). ε_{Nd}-values calculated using a decay constant for ^{147}Sm of 6.54 × 10^{-12} year^{-1} (Lugmair & Marti 1978) and CHUR parameters: $^{147}Sm/^{144}Nd$ = 0.1966 and $^{143}Nd/^{144}Nd$ = 0.512638 (Jacobsen & Wasserburg 1980; Wasserburg *et al.* 1981). Crustal array reference line ($\varepsilon_{Hf} = 2\varepsilon_{Nd} + 2$) after Vervoort & Patchett (1996).

Fig. 8.

Empirical cumulative distribution function plots of Natal Group samples.

Fig. 9.

Southern Gondwana at 480 Ma (after Torsvik & Cocks 2013) showing ε_{Hf} data from the Saldania Belt (Frimmel *et al.* 2013), Cape S-type granites (Villaros *et al.* 2012) Mozambique Belt (Thomas *et al.* 2010), Ecca Group (Veevers & Saeed 2007) and Dronning Maud Land (Veevers & Saeed 2007). For comparison, a 95th percentile contour of the Natal Group data has been added to each panel.

Table 1. *Sample coordinates and lithologies*

Sample	Geological unit	Lithology	Coordinates	
SA12/06	Newspaper Mbr	Sandstone	30°17'22.50"S	30°45'32.80"E
SA12/43	Newspaper Mbr	Sandstone	28°04'46.38"S	31°52'39.00"E
SA12/42	Tulini Mbr	Sandstone	28°05'00.66"S	31°52'39.06"E
SA12/20	Dassenhoek Mbr	Sandstone	29°48'21.01"S	30°48'13.95"E
SA12/27	Eshowe Mbr	Sandstone	28°34'41.04"S	31°19'10.74"E
SA12/39	Ulundi Mbr	Sandstone	28°05'09.00"S	31°52'32.46"E
SA12/29	Ulundi Mbr	Conglomerate matrix	28°32'38.16"S	31°08'29.16"E
SA12/28	Ulundi Mbr	Quartzite clast in conglomerate	28°32'38.16"S	31°08'29.16"E
SA12/24	Mgeni pluton	Granite	29°50'27.00"S	30°42'33.90"E
SA13/133	Fafa pluton	Granite	30°17'36.67"S	30°36'54.67"E

Table 2. *Results of one-dimensional likeness tests (Satkoski et al. 2013) comparing the Natal Group samples*

SA12/06	SA12/43	SA12/42	SA12/20	SA12/27	SA12/39	SA12/29	SA12/28
SA12/06	0.68	0.83	0.92	0.77	0.66	0.03	0.00
	SA12/43	0.74	0.68	0.74	0.72	0.13	0.08
		SA12/42	0.85	0.84	0.73	0.03	0.00
			SA12/20	0.77	0.68	0.03	0.00
				SA12/27	0.70	0.04	0.01
					SA12/39	0.14	0.11
						SA12/29	0.79
							SA12/28

Table 3. *Results of two-dimensional likeness tests (Satkoski et al. 2013) comparing the Natal Group samples*

SA12/06	SA12/43	SA12/42	SA12/20	SA12/27	SA12/39	SA12/29	SA12/28
SA12/06	0.57	0.71	0.82	0.70	0.53	0.00	0.00
	SA12/43	0.62	0.57	0.56	0.47	0.06	0.08
		SA12/42	0.72	0.68	0.54	0.00	0.00
			SA12/20	0.71	0.50	0.00	0.00
				SA12/27	0.60	0.01	0.01
					SA12/39	0.13	0.13
						SA12/29	0.81
							SA12/28

Table 4. *P-values for one-dimensional pairwise Kolmogorov-Smirnoff of the Natal Group samples*

SA12/06	SA12/43	SA12/42	SA12/20	SA12/27	SA12/39	SA12/29	SA12/28
SA12/06	0.00	0.12	0.88	0.11	0.00	0.00	0.00
	SA12/43	0.00	0.00	0.00	0.49	0.00	0.00
		SA12/42	0.25	0.78	0.00	0.00	0.00
			SA12/20	0.16	0.00	0.00	0.00
				SA12/27	0.00	0.00	0.00
					SA12/39	0.00	0.00

	SA12/29	0.00
		SA12/28

Table 5. *H-values for two-dimensional pairwise Kolmogorov-Smirnoff tests of the Natal Group samples*

SA12/06	SA12/43	SA12/42	SA12/20	SA12/27	SA12/39	SA12/29	SA12/28
SA12/06	1	0	0	0	1	1	1
	SA12/43	1	1	1	1	1	1
		SA12/42	0	0	1	1	1
			SA12/20	0	1	1	1
				SA12/27	1	1	1
					SA12/39	1	1
						SA12/29	0
							SA12/28

An H-value of 1 indicates that the null hypothesis – that both data sets were drawn from the same continuous distribution – should be rejected (i.e. $p < 0.05$), the reverse is true for H=0.

Table 6. *Results of one-dimensional likeness tests (Satkoski et al. 2013) comparing sedimentary sequences from southern Gondwana including Natal Group sandstones*

NGS	SB	KCCG	EG	DML	EWM	TMGEB	TMGWB
NGS	0.61	0.76	0.60	0.64	0.59	0.60	0.63
	SB	0.56	0.53	0.63	0.60	0.63	0.68
		KCCG	0.63	0.81	0.65	0.73	0.79
			EG	0.70	0.78	0.66	0.65
				DML	0.71	0.83	0.80
					EWM	0.72	0.70
						TMGEB	0.77
							TMGWB

NGS: Natal Group sandstones; SB: Saldania Belt (Frimmel *et al.* 2013); KCCG: Kansa and Cango Caves Groups (Naidoo *et al.* 2013); EG: Ecca Group (Veevers & Saeed 2007); DML: Dronning Maud Land (Veevers & Saeed 2007); EWM: Ellsworth-Whitmore Mountains (Flowerdew *et al.* 2007); TMGEB and TMGWB: Table Mountain Group Eastern and Western Basin, respectively (Fourie *et al.* 2010).

Table 7. *Results of two-dimensional likeness tests (Satkoski et al. 2013) comparing sedimentary sequences from southern Gondwana including Natal Group sandstones*

NGS	SB	EG	MB	DML	EWM	P	VS	CG
NGS	0.41	0.46	0.42	0.51	0.45	0.21	0.13	0.34
	SB	0.44	0.39	0.38	0.47	0.27	0.12	0.43
		EG	0.56	0.54	0.61	0.47	0.22	0.55
			MB	0.51	0.42	0.24	0.16	0.36
				DML	0.45	0.36	0.16	0.41
					EWM	0.46	0.29	0.56
						P	0.32	0.48
							VS	0.23
								CG

NGS: Natal Group sandstones; SB: Saldania Belt (Frimmel *et al.* 2013); EG: Ecca Group (Veevers & Saeed 2007); MB: Mozambique Belt (Thomas *et al.* 2010); Dronning Maud Land (Veevers & Saeed 2007); EWM: Ellsworth-Whitmore Mountains (Flowerdew *et al.* 2007); P: Patagonia (Uriz *et al.* 2010); VS: Ventania System (Argentina) (Ramos *et al.* 2014) CG: Cape Granite (S-type granite) (Villaros *et al.* 2012).