Crustal evolution of the amphibolite- to granulite-facies transition zone in the Eastern Dharwar craton, southern India: Insight from petrological modelling, zircon U–Pb geochronology, and Hf isotopes

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Abstract

Integrated petrological, geochemical, isotopic, and thermobarometric study of metasedimentary rocks from the amphibolite- to granulite-facies transition zone of the Eastern Dharwar Craton (EDC), south India, has provided new insight into the evolution of the lower continental crust in this region. Phase equilibrium modelling of metapelites and metagraywackes suggest that they reached peak metamorphism at ~800–850 °C and 6–7 kbar (corresponding to a paleodepth of ~20 km), with minor retrograde change occurring at ~700 °C and 3–5 kbar during exhumation. U–Pb ages of conventionally separated zircon from metapelite samples range from 2.5 to 3.4 Ga, whereas garnet-hosted zircons yield younger ages of 2.5–2.7 Ga. Zircon Th/U ratios and Hf isotopes reveal several significant pulses of zircon growth at 3.0, 2.95 and 2.7 Ga. Hf isotope data suggest evolution of juvenile magma at around 3.2 Ga, while Hf model ages show that crust building process also involved recycling of pre-existing Mesoarchean crust. Our study confirms the presence of a Paleoarchean component in the EDC lower crust, as well as older metamorphic events in the terrain and the gradational distribution of the metamorphic rocks.

KEYWORDS

Dharwar craton, juvenile magma, metapelite, petrological modelling, U–Pb geochronology, zircon Lu–Hf isotope

1. INTRODUCTION

Archean cratons commonly preserve evidence for multiple metamorphic, magmatic, and deformational events that occur over tens or hundreds of millions of years. Such events may be responsible for recycling of old crust and/or the accretion of juvenile material (e.g., Zeh, Gerdes, Klemd & Barton, 2007, 2008). Reconstruction of pressure–temperature–time (*P–T–t*) histories for key metamorphic lithologies in complex crustal sections represents a crucial step towards unravelling the tectonic processes that occurred on the early Earth. This can be facilitated by the application of key geochronometers, such as U–Pb in zircon, which can preserve evidence of multiple tectonothermal episodes (e.g., Rubatto, 2017). The behaviour of zircon during metamorphism of metasedimentary rocks is of critical importance, as U–Pb ages obtained from different grains may date inheritance, the timing of protolith formation, prograde, peak, or retrograde metamorphism, as well as intermediate episodes of sun-deformational partial melting and crystallization (e.g., Harley, Kelly, & Möller, 2007; Rubatto & Hermann, 2007; Yakymchuk & Brown, 2014; Yakymchuk, Clark, & White et al., 2017). The correct interpretation of such radiogenic ages in metamorphic rocks can be best achieved by combining such data with independent isotope systems, such as Lu–Hf, which can provide insight into the crustal processes that have affected a particular terrane.

Combined zircon U–Pb and Lu–Hf isotopic analyses can provide information about the timing of crustal differentiation or reworking events, and has been proven to be extremely useful in unravelling the magmatic–metamorphic history of complex polymetamorphic Archaean gneiss terranes (e.g., Gerdes & Zeh, 2009; Zeh et al., 2007). In particular, $^{176}Hf/^{177}Hf$ ratios in zircon can be used to trace zircon-forming metamorphic reactions in high-grade rocks. Thus, collecting U–Pb and Lu–Hf isotope data from individual zircon grains can provide high-resolution information about the timing and nature of metamorphism and magmatism in a sample.

Previous study of zircon in rocks from the Dharwar craton, India, has revealed multiple metamorphic episodes, with Jayananda et al. (2013) documenting heating/prograde events at ~3.2, 2.62, and 2.55-2.52 Ga, followed by cooling until 2.45 Ga. Other studies have reported the possibility of an additional metamorphic event at ~3.0 Ga (Mahabaleshwar and Peucat, 1988; Mahabaleshwar et al., 2013). Nonetheless, the timing of metamorphism across the amphibolite-granulite transition zone in the Eastern Dharwar Craton (EDC) is uncertain. In this study, U–Pb ages and Lu–Hf isotope data from zircon grains are combined with detailed petrographic observations and phase equilibrium modelling of metapelitic rocks from the amphibolite–granulite transition zone in the EDC. Together, these data constrain the timing and *P*–*T* conditions of high-grade metamorphism as well as post-peak metamorphic processes, and relate the metamorphic evolution to previously reported regional metamorphic events. These results provide new insight into the processes and timescales over which thickened continental crust in southern India has been deformed.

2. GEOLOGICAL BACKGROUND

The Dharwar craton basement consists primarily of a granite–greenstone terrain in the north and a granulite-facies gneiss terrain in the south (Figure 1). The craton has historically been divided into two parts – the Western Dharwar craton (WDC) and the Eastern Dharwar craton (EDC) – based on the greenstone belt characteristics, degree of metamorphism and age, nature of the gneissic rocks and crustal thickness (e.g., Swami Nath & Ramakrishnan 1981; Radhakrishna & Vaidyanathan 1997; Jayananda et al. 2000; Gupta et al. 2003; Ramakrishnan & Vaidyanathan 2008), although recent workers have identified a putative ‘central’ section that has a discrete tectonic history (cf. Li et al. 2019). The greenstone-granite gneiss terrain consists of N–S to NNW–SSE trending supracrustal belts, known as the Dharwar schists, which are separated from each other by gneisses, known as the Peninsular Gneiss. The supracrustal sequence in the Dharwar schists also occurs as enclaves in Peninsular Gneiss, and has been classified into two lithostratigraphic divisions: the older Sargur Group and the younger Dharwar Supergroup (Swami Nath & Ramakrishnan, 1981). The supracrustal rocks of the greenstone belts and the gneisses grade into a granulite complex in the southern part of the craton. In the EDC, there are sericitic and fuchsitic quartzites, metapelites (andalusite, corundum-bearing or cordierite–orthopyroxene–sillimanite-bearing schists and gneisses), calc-silicate rocks and banded manganiferous iron formations occurring as enclaves in orthogneisses. These have been correlated with the Sargur Group of the WDC (Swami Nath & Ramakrishnan, 1981). Zircon U–Pb and Lu–Hf systematics for the EDC show that some lithologies are older than 3 Ga (Maibam et al., 2011), and some others have protolith ages as old as c. 3.5 Ga (Maibam et al., 2016). Crustal reworking in the EDC can be dated back to 3.3 Ga (Maibam et al., 2016).

The EDC preserves a continuous increase in metamorphic grade from greenschist-facies in the north, through amphibolite-facies rocks in the centre, to granulite-facies rocks in the south (Pichamuthu, 1965; Raith, Rasse, Ackermand & Lal, 1982; Raase, Raith, Ackermand, & Lal, 1986). Although metapelitic rocks are widespread in each facies, few studies have focused on the petrological changes that occur across this amphibolite-to-granulite transition, where partial melting occurs. It remains unknown whether this southward increase in metamorphic grade is related to a single thermal event or multiple thermal events (Jayananda, Tsutsumi, Miyazaki, Gireesh, Kapfo, Tushipokla, Hidaka, & Kano, 2013). Published geochronological and structural data indicate a craton-wide thermal event that affected all of the Dharwar Archean crust from 2.55 to 2.51 Ga (Friend and Nutman, 1991, 1992; Chardon, Jayananda, Chetty, & Peucat, 2008, Maibam, Goswami & Srinivasan, 2011; Peucat, Mahabaleswar & Jayananda, 1993; Peucat, Jayananda, Chardon, Capdevila, Fanning, & Paquette, 2013), although an earlier episode of metamorphism at c. 3.0 Ga has also been suggested (Mahabaleswar & Peucat, 1988; Mahabaleswar, Jayananda, Peucat, & Swamy, 1995). Jayananda, Banerjee, Pant, Dasgupta, Kano, Mahesha, & Mahabaleswar (2012) documented 2.62 Ga granulite-facies assemblages close to ultra-high temperature (UHT) conditions in the central part of the eastern Dharwar craton. All of these studies suggest that more than one regional-scale, high-grade metamorphic episode occurred in the Dharwar craton. Here, we integrate petrological and phase equilibria modelling with geochronology and Lu–Hf isotopic analysis of metasedimentary zircon to examine the relationships between magmatism and metamorphism in this region of India.

3. SAMPLE PETROLOGY AND MINERAL COMPOSITIONS

The EDC mainly contains high-grade supracrustal rocks, amphibolite facies gneisses, charnockites, mafic granulites, and pink K-feldspar rich granites. The high-grade supracrustal rocks form discontinuous enclaves containing quartzite, metapelite, calc silicate rocks and banded iron formations, and are considered stratigraphic equivalents of the Sargur Group in the WDC (Ramakrishnan & Viswanatha, 1981).

We collected 30 metapelite samples from the EDC, encompassing all metamorphic grades. Three amphibolite- and granulite-facies samples (ED3, ED6, and ED8) were chosen for thermobarometry. To constrain the timing of polymetamorphic events, we selected ~~two~~ sample ED6, belonging to the amphibolite facies and ED8, belonging to the granulite facies ~~were analyzed~~ metamorphism for U–Pb zircon geochronolog~~y~~ical study with ED6 as a subject for Lu–Hf isotopic study due to low age dispersion ~~work~~. Sample ED6 is a cordierite-bearing metapelite collected from a location about 100 meters west of Honniganahatti village (N12°58′34.0″, E77°25′01.4″), ED8 is a metagraywacke sample from the west of the Bangaluru-Mysore Highway between Ramanagaram and Channayapatna (N12°42′12.6″, E77°15′13.1″), and ED3 is a high-Mg metapelite from the road cut near the Narasandra village along the Bangaluru-Mangalore Highway (N13°03′45.5″, E77°09′39.9″). Sample locations are shown on figure 1. We also used selected thin sections of garnet-bearing metasedimentary samples for *in situ* U–Pb geochronological analysis of zircon inclusions in garnet porphyroblasts.

Samples ED3 and ED6 are migmatitic metapelites, the former being more magnesian than the latter (Supplementary Table 2). This compositional difference is likely due both samples having different protoliths (see below). Both are characterized by discontinuous quartzofeldspathic leucosome segregations that occur in thick, weakly foliated (ED3) and unfoliated (ED6) host rock. Leucosomes contain quartz, plagioclase, K-feldspar, minor biotite, alongside porphyroblasts of orthopyroxene (ED3), or garnet and cordierite (ED6). The host rock in each is dominated by quartz, plagioclase, and biotite, with additional orthopyroxene and magnetite in ED3, and minor sillimanite, garnet, and cordierite in ED6. Accessory monazite, zircon, and apatite occur in each. Orthopyroxene in ED3 forms coarse, subhedral to anhedral grains up to 1 cm in diameter, which are compositionally unzoned, with X_{Mg} = Mg/(Fe + Mg) cations per formula unit (cpfu) = 0.55–0.57. These porphyroblasts typically contain globular quartz inclusions and are wrapped by biotite (Figure 2b). Biotite has similar compositions in both the leucosome and host rock, with X_{Mg} = 0.51–0.55 and Ti = 0.20–0.25 (for 11 oxygens), and cordierite has X_{Mg} = 0.75–0.77. Plagioclase has X_{An} = Ca/(Ca + Na + K) cpfu = 0.38–0.40 (Supplementary Table 1). Garnet porphyroblasts in ED6 show no systematic compositional variation within or between grains ($Alm_{65–70}Prp_{18–23}Grs_{4–6}Sps_{2–3}$), except for minor Mn-enrichments at their outermost rims (up to 5%). Neither biotite nor cordierite shows internal zonation or consistent compositional variation between leucosome and matrix domains, with both exhibiting X_{Mg} = 0.54–0.63 and 0.65–0.72, respectively (Supplementary Table 1). Plagioclase in ED6 has X_{An} = 0.39–0.42 (Supplementary Table 1) and shows ~120° angles at triple junctions (Figure 2c), indicative of textural equilibration at high temperatures. Myrmekite also occurs in leucosome domains (Figure 2c).

Metagreywacke sample ED8 is coarse-grained, massive, and contains garnet, cordierite, biotite, plagioclase, K-feldspar, ilmenite, and quartz. Accessory apatite and zircon also occur. Only minor evidence of partial melting is visible at the thin section scale as isolated melt pockets and films of crystallized melt against garnet (Figure 2d, red box). Garnet occurs as cm-scale porphyroblasts that are compositionally homogeneous ($Alm_{62–70}Prp_{21–27}Grs_{2–3}Sps_3$). Plagioclase and K-feldspar in ED6 have X_{An} = 0.22–0.27 and ~0.01, respectively. Cordierite occurs as subhedral porphyroblasts that have X_{Mg} = 0.74–0.75, and biotite occurs as unaligned flakes with X_{Mg} = 0.50–0.55 and Ti = 0.13–0.27 (for 11 oxygens).

4. ANALYTICAL TECHNIQUES

Mineral compositional data for all samples were obtained on a JEOL JXA-8200 electron microprobe housed at the Institute of Geosciences, Johannes-Gutenberg University of Mainz, Germany. Operating conditions included an acceleration voltage of 15 kV, a beam current of 12 nA, and a 2-μm spot size. A matrix correction for atomic number, absorption, and fluorescence was automatically applied to all analyses. For the data presented below, mineral compositions were recalculated to a standard numbers of oxygens per formula unit (pfu) using AX (Holland 2009), with H_2O assumed to be present in stoichiometric amounts. Representative compositions of major minerals are given in Supplementary Table 1.

Bulk-rock compositions were obtained via X-ray fluorescence (XRF) via the production of glass pellets in order to guarantee standardized and reproducible analyses. Powdered rock samples were initially dried overnight at 105 °C. Approximately 5.2 g of lithium tetraborate ($Li_2B_4O_7$) flux and 0.4 g of powdered rock sample were then weighed, homogenized, and melted in a Vulcan AMA melting device to produce each glass pellet. These pellets were then analysed by a Philips MagXPRO spectrometer**,** with a rhenium X-ray tube at the Institute of Geoscience, Johannes Gutenberg University of Mainz, Germany. Detection limits are estimated to be 100 ppm for light elements (Na, Mg, Al) and 10 ppm for heavy elements (K to U). Analysed major oxides comprised SiO_2, Al_2O_3, total Fe_2O_3, MnO, MgO, CaO, Na_2O, K_2O, TiO_2, P_2O_5, SO_3, Cr_2O_3, and NiO. These data are presented in Supplementary Table 2.

4.1. Zircon geochronology

Zircon from metapelite samples were analysed via grain mounts of separated grain fractions, along with in-situ analysis of zircon inclusions in garnet porphyroblasts in polished thin sections. The samples were crushed into centimetre-sized chips and were thoroughly washed after eliminating the weathered portions. The clean chips were pulverized to <250 μm using a stainless-steel piston and cylinder. After repeated washing, non-magnetic, high-density mineral grains were concentrated by density separation using aqueous sodium polytungstate solution (density = 3 g/cm^3) followed by magnetic separation using a Frantz isodynamic separator. Clear, unfractured, zircon grains were selected and were mounted on an adhesive double-sided tape, cast in epoxy and sectioned by polishing. Transparent zircons with simple internal structure were documented in detail. Most grains are subhedral to anhedral and rounded, or weakly elongated prisms with rounded tips (Supplementary Figure 1). The grains recovered from the studied samples are inclusion free, have elongate or rounded forms, are colourless to brown in colour, and most core domains are metamict. The tips of the grains are mostly rounded. Few zircons show distinct overgrowths; however, these rims could not be analysed due to their small size. Cathodoluminescence (CL), back scattered electron images (BSE) of separated zircons, and representative zircon grains occurring as inclusions in garnet porphyroblasts are presented in Supplementary Figure 1.

Uranium, thorium and lead isotopes for ED6 zircon were analysed using a ThermoScientific Element 2 sector field (SF) ICP-MS coupled to a RESOlution M-50 (Resonetics) 193 nm ArF excimer laser (CompexPro 102, Coherent) system equipped with an S-155 two-volume ablation cell (Laurin Technic, Australia) at Goethe-University Frankfurt (GUF). Laser spot-size was 23 μm for unknowns and zircon reference material (RM) GJ1, 91500, and OG. Sample surface was cleaned directly before each analysis by a four pulses pre-ablation. Ablation was performed in a 0.3 L min^{-1} He stream, which was mixed directly after the ablation cell with 0.007 L min^{-1} N_2 and 0.9 L min^{-1}Ar prior to introduction into the Ar plasma of the SF-ICP-MS. The gases all 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}UO/^{238}U, below 0.5%. The sensitivity achieved was in the range of 8000–12,000 cps/μg g^{-1} for ^{238}U with a 23 μm spot size, at 5.5 Hz and 5–6 J cm^{-2}. The typical penetration depth was about 15–18μm. The two-volume ablation cell S-155 enables detection and sequential sampling of heterogeneous grains (for example growth zones) during time resolved data acquisition, due to its quick response time of <1 s (time until maximum signal strength was achieved) and wash-out (<99% of previous signal) time of <3 s. During the time-resolved processing contamination resulting from inclusions and compositional zoning was monitored and only the relevant part of the signal was integrated. 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 & Zeh, 2006, 2009). Common Pb correction have been applied based on the ^{208}Pb signal, corrected for radiogenic ^{208}Pb following the method described in Millonig, Gerdes & Groat (2013). For the analysed samples, the ^{204}Pb content was mostly near or below the detection limit (see supplementary table 3A). Laser-induced elemental fractionation and instrumental mass discrimination were corrected by normalization to the RM GJ-1 (Jackson, Pearson, Griffin & Belousova, 2004). Prior to this normalization, the inter-elemental fractionation (^{206}Pb/^{238}U) during the 21 s of sample ablation was corrected for each individual analysis. The correction was done by applying a linear regression through all measured ratios, excluding the outliers (±2 standard deviation; 2 SD), 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 (0.0982 ± 0.0004; ID-TIMS GUF value) of the analysed GJ-1 grain was around 6% and the drift over the day was less than 2%.

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 (n = 10; 2 SD ~1.8%) during the analytical session (~4 h) and the within-run precision of each analysis (2 SE; standard error). For $^{207}Pb/^{206}Pb$ we adopt a ^{207}Pb signal-dependent uncertainty propagation (see Gerdes & 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 modern $^{238}U/^{235}U$ natural abundance ratio of 137.88 and the uncertainty derived by quadratic addition of the propagated uncertainties of both ratios. The accuracy of the method was verified by analyses of reference zircon 91500 (1060.6 ± 9.0 Ma, MSWD of concordance and equivalence ($MSWD_{C+E}$) = 0.81, n = 5), and OG (3463.6 ± 8.8 Ma, $MSWD_{C+E}$ = 0.49, n = 5). The data were plotted using the software ISOPLOT (Ludwig, 2003).

U–Pb isotopic analyses of the garnet-hosted zircons and sample ED8 were performed at the Westfälische Wilhelms-Universität Münster, Germany with an Element 2 mass spectrometer (ThermoFisher) connected to a Photon Machines Analyte G2. Forward power was 1300 W and reflected power <1 W, gas flow rates were 1.2 l/m for He (carrier gas of ablated material), 0.9 l/m and 1.1 l/m for the Ar-auxiliary and sample gas, respectively. Cooling gas flow rate was set to 16 l/min. Before starting analysis, the laser repetition rate was set to 10 Hz using a fluence of 4 J/cm^2 and the system was tuned (torch position, lenses, gas flows) on a NIST 612 glass measuring ^{139}La, ^{232}Th and $^{232}Th^{16}O$ to get stable signals and high sensitivity, as well as low oxide rates ($^{232}Th^{16}O/^{232}Th$ < 0.1%) during ablation. Laser spot sizes varied between 25 and 45 µm, depending on the size of the zircons. Masses ^{202}Hg, $^{204}Hg/Pb$, ^{206}Pb, ^{207}Pb were measured in e-scan mode with four samples per peak except for ^{238}U

which was measured with 10 samples per peak. Dwell time was 40 ms for ^{202}Hg, $^{204}Hg/Pb$, and ^{207}Pb, 10 ms for ^{206}Pb and 4 ms for ^{238}U. Overall time of a single analysis was 64 s (12 s for background, 37 s for peak after switching laser on, 15 s washout). Groups of ten unknowns were bracketed with three analyses of the GJ-1 reference zircon (Jackson et al. 2004) for external calibration. Measurements (n = 21) of the 91500 standard zircon (Wiedenbeck, Allé, Corfu, Griffin, Meier, Oberli, von Quadt, Roddick, & Spiegel, 1995) as unknowns during the analytical session yielded values of 0.1797 (± 2.9 %, 2σ) for the $^{206}Pb/^{238}U$, 1.858 (± 3.8 %, 2σ) for $^{207}Pb/^{235}U$, and 0.0750 (± 2.5 %, 2σ) for $^{207}Pb/^{206}Pb$ ratios, and match published values (Wiedenbeck et al., 1995) within error. The U contents were estimated relative to the GJ-1 reference. Data reduction was performed with an in-house Microsoft Excel spreadsheet. Concordia diagrams were constructed using Isoplot3.00 (Ludwig, 2003). Further details of the analytical and data reduction protocols can be found in Kooijman, Berndt & Mezger (2012).

4.2. Lu–Hf–Yb Isotope Analyses

Hafnium isotope measurements were performed with a Thermo-Finnigan Neptune multicollector (MC) ICP-MS at GUF coupled to the same laser as described in the U–Pb method (RESOlution 193 nm ArF excimer laser). Spots with 40-μm diameters were drilled with a repetition rate of 5.5 Hz and a fluence of 6 J/cm^2. Data were collected in static mode (^{172}Yb, ^{173}Yb, ^{175}Lu, ^{176}Hf–Yb–Lu, ^{177}Hf, ^{178}Hf, ^{179}Hf, ^{180}Hf) during 40 seconds of laser ablation per spot analysis. All data were reported relative to the JMC475 $^{176}Hf/^{177}Hf$ ratio of 0.282160 and quoted uncertainties are quadratic additions of the within-run precision of each analysis and the JMC475 repeatability (2 S.D. = 0.0033%, n = 8). Accuracy and external reproducibility of the method was verified by repeated analyses of reference zircon GJ-1 and

Plešovice, which yielded $^{176}Hf/^{177}Hf$ values of 0.282001 ± 0.000019 (2 S.D., n = 6) and 0.282481 ± 0.000022 (n = 32), respectively. This is well within the range of solution mode data (Sláma, Košler, Condon, Crowley, Gerdes, Hanchar, Horstwood, Morris, Nasdala, Norberg, Schaltegger, Schoene, Turbett & Whitehouse, 2008; Gerdes & Zeh, 2006).

For calculation of the epsilon Hf [εHf_t], the chondritic uniform reservoir (CHUR) as recommended by Bouvier, Vervoort & Patchett (2008; $^{176}Lu/^{177}Hf$ = 0.0336 and $^{176}Hf/^{177}Hf$ = 0.282785), and a decay constant of 1.867 × 10^{-11} were used (average of Scherer, Münker & Mezger, 2001; Söderlund, Patchett, Vervoort & Isachsen, 2004). Initial $^{176}Hf/^{177}Hf_t$ and εHf_t for all analysed zircon domains were calculated using the apparent Pb–Pb ages obtained, and for all co-genetic zircon domains by using the intrusion ages of the respective granitoids. Depleted mantle hafnium model ages (T_{DM}) were calculated using values for the depleted mantle as suggested by Blichert-Toft and Puchtel (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 $\varepsilon Hf_{4.558Ga}$ = 0.0. 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, and $^{176}Lu/^{177}Hf$ =0.01 for Paleoproterozoic–Archean crust (i.e., the mean of average continental crust; Taylor & McLennan, 1985; Wedepohl, 1995).

5. RESULTS OF ISOTOPIC STUDY OF ZIRCON

5.1. U–Pb geochronology

CL and BSE images show complex internal structure for most of the analysed grains (Supplementary Figure 1). Some of the grains show rounded grain morphology. Many have clear core–rim structures and oscillatory zoning, while a few grains are homogenous. U–Pb isotope analyses were conducted on 51 zircon grains from metapelite sample (ED6) and 23 grains from ED8, and the data are presented in Supplementary Table 3. Most of the grains show medium to high U concentration (337 to 1781 ppm). Only analyses with degrees of concordance between 90% and 105% were considered for age determination (Supplementary Figure 2). Five distinct age populations are identified, with five grains (A115, A135, A141, A144-145) having ages between 3.2 Ga and 3.4 Ga (Th/U = 0.13–0.48) and ten grains (A100, A107-109, A111, A121, A128, A133, A143, A155) with ages ~3.0 Ga (Th/U = 0.002 to 0.079) and they yielded concordant ages (concordance level 95–102; Supplementary Figure 2). Two younger ages at ~2.7 Ga (A116, A118) (Th/U = 0.002–0.003) and one at 2.5 Ga (A137) (Th/U = 0.002) were also found. A probability distribution diagram is presented in Figure 3. Discordant analyses (A119, A120, A132, A140, A151) are interpreted to represent either single or multiple partial Pb-loss events. It may be noted that $^{207}Pb/^{206}Pb$ ages of the discordant analyses are similar to the reported age spectrum, suggesting recent Pb-loss event. Heterogeneous zircon grains with distinct core–rim morphologies from samples from the same region of the Dharwar craton, with core ages of ~3.35–3.00 Ga, and, rims with ages of ~2.75–2.70 Ga have been reported earlier (Maibam et al., 2011).

Measured U and Th ~~concentrations~~ ratios in zircon and grain morphology and internal texture were used to discriminate between grains (or domains within grains) that had a magmatic or metamorphic origin (cf. Hoskin & Schaltegger, 2003; Hoskin and Black, 2003, Rubatto, 2017; Schaltegger, Fanning, Günther, Maurin, Schulmann, & Gebauer, 1999; Williams & Claesson, 1987). In most geological environments, metamorphic zircons have Th/U ratios <0.1 (Rubatto, 2017) whereas most unaltered magmatic zircons have Th/U ratios >0.1 (Belousova, Griffin, O'Reilly, & Fisher, 2002; Grimes, Wooden, Cheadle & John, 2015). In sample ED6, zircon younger than c. 3.1 Ga has uniformly low Th/U ratios (0.002 to 0.079), whereas zircon older than c. 3.2 Ga has high Th/U values (>0.1) characteristic of a magmatic origin. These results indicate that older magmatic (>3.2 Ga) rocks have been overprinted by a younger metamorphic episode.

Twenty-three analyses on 21 zircon grains from sample ED8 (Supplementary Table 3b (Supplementary Figure 2b)) show medium to high U concentrations (127 to 1451 ppm). The dataset yielded concordant ages (96–103% concordance) ranging from 3.4 to 2.5 Ga, with a dominant age group between 3.1 and 2.7 Ga. Four age populations are observed, with five grains having age older than 3.3 Ga, and three grains yielding age ~3.0 Ga, eight analyses ranging between 2.8 to 2.9 Ga and seven analyses between 2.8 to 2.7 Ga and two analyses with ~2.5 Ga. The U–Pb dataset is presented in Supplementary Table 3 and the concordia and probability distribution diagrams in Supplementary Figure 2 and Figure 3 respectively. Heterogeneous zircon grains with distinct core-rim morphology showing with core age of ~2.77 Ga and rim with ~2.51 Ga. Older ages obtained for the other grains are higher than the radiometric ages of ~3 Ga reported previously for samples from this region (Mojzsis, Devaraju & Newton, 2003; Mahabaleswar et al., 1995; Friend & Nutman, 1991) except for zircon and monazite ages of 3.2 Ga reported by Jayananda et al. (2013).

Inclusions of zircon in metapelitic garnet from the two samples (ED6 and ED8) were analysed in situ for U–Pb geochronology (Supplementary Table 3b). The analysed grains show a wider U concentration (68 to 2046 ppm). Fourteen analyses in 13 zircons were carried out and five of these analyses were discordant (<91%). The analysed ages for the other zircons range between 2.74 to 2.54 Ga, with the core of one grain yielding an age of 3.1 Ga and the rim being 2.6 Ga. Three analyses give ages of >2.55 Ga, five analysis yielded ages ranging between ~2.6 to 2.65 Ga, and two analyses yielded an age of ~2.75 Ga. Calculated $^{207}Pb/^{206}Pb$ ages of the discordant analyses are similar to the reported age spectrum, indicative of recent Pb-loss event. A probability distribution diagram for these data is presented in Figure 3b. The core–rim ages in the heterogeneous zircon grain are compatible with the reported core–rim ages of the monazite from the area (Jayananda et al., 2013). The zircon grains yielding younger ages (2.5 to 2.6 Ga) are small and some may have been lost during mineral separation.

5.2. Lu–Hf isotope

Sample ED6 was selected for Lu–Hf isotopic analysis due to the low age dispersion. Forty-five Lu–Hf isotopic analyses were conducted on zircon grains of sufficient size (Supplementary Table 4), which representing all four age groups (Figure 4). All but one grain in the first group of magmatic zircons (older than 3.2 Ga) yielded chondritic to superchondritic εHf of +0.5 to +2.9, corresponding to Hf model ages of 3.22 to 3.36 Ga and a $^{176}Hf/^{177}Hf$ ratio of 0.280632 to 0.280755. The second group of seven grains with Pb–Pb ages of ~3.1 Ga have $^{176}Hf/^{177}Hf$ of 0.280692 to 0.280785, εHf(*t*) of −1.6 to −4.8, and T_{DM} of 3.37–3.47 Ga. The third group yielding Pb–Pb ages of 2.9–3.0 Ga have $^{176}Hf/^{177}Hf$ of 0.280668 to 0.280809, εHf(t) of −4.2 to −11, and T_{DM} of 3.37 to 3.69 Ga. The fourth group yielding Pb–Pb ages of 2.7–2.6 Ga have $^{176}Hf/^{177}Hf$ of 0.280708 to 0.280738, εHf(t) of −10.8 to −12.2, and T_{DM} 3.51 to 3.56 Ga. The two youngest grains (~2.5 Ga) have $^{176}Hf/^{177}Hf$ of 0.280918–0.280770, εHf of-7.3 to -16.2, and T_{DM} of 3.16 to 3.65 Ga (Figure 4). However, it may be noted that depleted mantle model ages commonly not represent ages of geologic events but are numerical estimates of average mantle extraction ages (Spencer, Kirkland, Roberts, Evans, & Liebmann, 2020) and should not be used to constrain quantitative chronologic information.

The analysed zircons show a negative correlation between εHf(t) and Pb–Pb age (Figure 4), but most show very similar initial $^{176}Hf/^{177}Hf$ values (Figure 4). We interpret this to be due to Pb-loss or resetting of the U–Pb system due to solid state recrystallization of zircon, which changes the U–Pb age but not the Hf isotope composition (Gerdes & Zeh, 2009). In contrast, new zircon that forms in response to metamorphism typically forms overgrowth domains with high $^{176}Hf/^{177}Hf$ and thus more positive εHf(t) (Liu, Gerdes, Zeng & Xue, 2008). Interpreting the U–Pb ages as magmatic events, the trend identified here corresponds to recycling of the 3.2-3.4 Ga juvenile crust between 3.1 and 2.7 Ga.

Interpretation of Hf isotope arrays through time generally uses an isotope trajectory defined by fix Lu/Hf ratio of a source and which then defines a $^{176}Hf/^{177}Hf$ line in isotope space. We used present day Archaean-Paleoproterozoic Lu/Hf value of 0.0113 (Taylor & McLennan, 1985; Wedepohl, 1995) to define the $^{176}Hf/^{177}Hf$ line. The combined U-Pb-Lu-Hf datasets from the studied metapelite dataset with a steady decrease of εHf(t) from + 2.9 to -16.2 between 3.45 to 2.52 Ga. It seems most likely that the younger (2.5-2.7 Ga) zircons formed by reworking of the 3.3 to 3.5 Ga rocks.

6. PETROLOGICAL MODELLING

Petrological modelling was performed to provide new constraints on the *P–T* conditions of metamorphism experienced by each of the studied rock types. All calculations were performed using THERMOCALC version 3.40 (Powell & Holland, 1988) with the internally consistent thermodynamic dataset ds62 (Holland and Powell, 2011) in the MnO–Na_2O–CaO–K_2O–FeO–MgO–Al_2O_3–SiO_2–H_2O–TiO_2–O (MnNCKFMASHTO) compositional system using the following *a–x* relations: garnet, silicate melt, biotite, muscovite–paragonite, cordierite, orthopyroxene, staurolite, chlorite, magnetite–spinel, ilmenite–hematite (White, Powell, Holland, Johnson & Green, 2014a; White, Powell & Johnson, 2014b), and plagioclase and K-feldspar (Holland & Powell, 2003). Andalusite, sillimanite, kyanite, rutile, quartz, and H_2O were treated as pure phases.

As XRF cannot discriminate between ions of different valence states, the molar bulk-rock XFe^{3+} (= Fe_2O_3/(FeO+Fe_2O_3)) ratios used for modelling of each sample were fixed at 0.1, owing to the minor proportion of ferric iron in recalculated mineral compositional analyses (Supplementary Table 1) and the absence (or <<1% proportion) of any key Fe^{3+}-bearing phases, such as epidote, magnetite, and hematite (cf. Diener & Powell, 2010). Modelled water contents were calculated from XRF-derived loss on ignition (LOI). Bulk compositions used for phase diagram construction are given in Supplementary Table 2. Mineral proportions for each sample were determined using the software JMicroVision (Roduit, 2010), with each individual count consisting of five hundred points randomly distributed over a digitally scanned thin-section image.

P–T pseudosections for samples ED3, ED6, and ED8 calculated between 2 and 10 kbar, and 650 and 950 °C, are shown in Figures 5a, 5b, and 5c, respectively. The terminal textural development of partially melted rocks commonly occurs upon crossing the solidus during retrograde cooling and exhumation (e.g., White & Powell, 2002). The calculated position of the solidus for each of the studied samples therefore provides a minimum temperature for peak metamorphism. The measured H_2O contents for samples ED3 (high-Mg metapelite) and ED6 (metapelite) were insufficient to saturate their solidi across the *P–T* range investigated, which occur at elevated temperatures up to 750 °C (Figure 5a) and 810 °C (Figure 5b) at low-pressure conditions, respectively. The interpreted peak (or near-peak) assemblage pl–bi–opx–cd–mt–ilm–q–liq (±H_2O) identified in sample ED3 is calculated to be stable at 730–850 °C at pressures below 5 kbar (Figure 6a; blue field, bold text), whereas the assemblage g–cd–pl–bi–sill–ilm–q–liq (±H_2O) is stable at 6–7 kbar and 710–840 °C (Figure 5b, green field, bold text). In the former case, higher temperature conditions than those that delimit the observed assemblage are calculated to destabilize plagioclase, and higher pressures would stabilize garnet (which is not observed in sample ED3), and the low-temperature limit is defined by the solidus. In the latter case, the peak assemblage field is bounded at higher-temperature–low-pressure conditions by biotite- and/or sillimanite-absent fields, and by cordierite-absent fields at higher pressures. As many of these limiting assemblage field boundaries involve the loss of key phases that are observed to be abundant in each sample, they provide tight constraints on the *P*–*T* conditions at which each rock equilibrated.

At *P*–*T* conditions of 4.75 kbar and 825 °C, towards the upper limit of the calculated peak assemblage field for sample ED3, sample ED3 would be comprised of ~33% quartz, ~32% cordierite, ~18% biotite, ~9% orthopyroxene, ~6% melt, and <1% plagioclase and ilmenite. These proportions differ slightly from observed proportions of ferromagnesian phases. At lower-grade conditions of 3 kbar and 750 °C, sample ED3 would be comprised of ~35% quartz, ~33% cordierite, ~20% biotite, ~6% orthopyroxene, ~2% melt, ~1% plagioclase, and <1% ilmenite and magnetite. For metapelite sample ED6, calculated phase proportions at *P*–*T* conditions of 6.5 kbar and 800 °C (just above the solidus) are ~22% garnet, ~21% quartz, ~21% plagioclase, ~16% biotite, ~9% cordierite, ~8% sillimanite, ~2% melt, and <1% ilmenite, which matches most closely those observed in the sample.

The measured H_2O content for metagreywacke sample ED8 is just sufficient to saturate the calculated solidus across the *P*–*T* range investigated, which is situated at 670–720 °C (Figure 5c) and so places a lower bound on the conditions of peak metamorphism. The interpreted peak (or near-peak) assemblage of g–pl–bi–cd–ilm–q–liq (±H_2O) is calculated to be stable over a wide range of *P*–*T* conditions between ~3.0–6.5 kbar and ~700–850 °C (Figure 5c; pink field, bold text), being bound at low pressure by the disappearance of biotite and stabilization of magnetite, and at high temperatures and pressures by the stabilization of orthopyroxene and sillimanite, respectively; neither of which was observed in the sample. At *P*–*T* conditions of 6 kbar and 825 °C, at the uppermost end of the interpreted peak assemblage field, calculated phase proportions comprised ~38% quartz, ~30% plagioclase, ~15% melt, ~8% garnet, ~6% cordierite, ~2% biotite, and <1% ilmenite; at 4.5 kbar and 710 °C, just above the solidus, calculated phase proportions comprised ~42% quartz, ~32%

plagioclase, ~10% cordierite, ~10% biotite, ~4% melt, <1% garnet, and <1% ilmenite. Thus, cooling and exhumation across this melt-bearing assemblage field is calculated to be associated with melt crystallization and the replacement of garnet by biotite and cordierite. Based on the observed phase proportions and textural relations in sample ED8, we interpret peak conditions to have been between ~800–850 °C at 6–7 kbar, with minor retrograde change occurring during cooling down-temperature until crossing the solidus at ~700 °C and 3–5 kbar (Figure 5d).

The metamorphic conditions of samples collected from the study area are relatively higher grade than those described previously (Harris and Jayaram, 1982). ~~These conditions lie on the granulite facies typical of Archaean metamorphism in the continental crust (e.g., White et al. 2017).~~ We interpret that the relatively low peak *P-T* conditions recorded by Harris and Jayaram (1982) can be attributed to previous workers having employed cation-exchange based conventional thermobarometry to determine conditions of equilibration. This approach is severely affected by cation diffusion between minerals that affects compositions during retrograde cooling (Frost and Chacko 1989; Kohn and Spear 2000). However, phase equilibrium modelling, as employed here, ~~represents an alternative approached by which to interprete the thermal history of metamorphic rocks, as it~~ relies primarily on the preserved mineral assemblage rather than mineral compositions (Powell and Holland, 2008) and allows the *P-T* path of metamorphism to be deduced from observed or interpreted changes in mineral compositions (e.g., Stüwe and Powell, 1995; White et al., 2002). The formulation and expansion of internally consistent thermodynamic datasets containing phases relevant to petrological investigations (e.g. Berman 1988; Holland and Powell 1998, 2011) and continued development of a-x relations for minerals and melts in suprasolidus lithologies (e.g. Green et al. 2016) has led to petrological modelling becoming the most reliable technique that can be used to address the thermobarometric evolution of metamorphic rocks, and constrain the tectonic processes that occurred during the ancient orogeny.

7. DISCUSSION AND CONCLUSIONS

Most published U-Pb and Pb-Pb ages of zircon and titanite from the EDC volcanic strata, gneisses and granite units span the range from 2.5 to 2.95 Ga (e.g., Krogstad, Hanson & Rajamani, 1991; Balakrishnan, Rajaman, & Hanson, 1999; Jayananda, Moyen, Martin, Peucat, Auvray & Mahabalesawar, 2000; Mojzsis et al., 2003). However, data obtained in this study for zircons in EDC metapelite enclaves record older age components ranging from 2.9 to 3.4 Ga. We note that ages between 2.7 to 2.8 Ga recorded in a few zircons (Supplementary Table 3) are similar to metamorphic ages reported for the EDC by Mojzsis et al. (2003). The scatter in the age data most probably reflects the complex succession of geological events in the EDC. Despite the poor state of preservation of the older gneissic rocks in the EDC, our data support magmatism having occurred in this region at 3.1-3.4 Ga (e.g., Maibam et al., 2016); however, we note that juvenile zircon ages of coeval age range have also been reported from the WDC (Maibam et al., 2016; Guitreau, Mukasa, Loudin & Krishnan, 2017).

Zircon ages obtained in this study lie within the range of previously reported ages of metasedimentary samples from the EDC, but are dominantly >3.0 Ga. Ages ranging from ~2.9 to 3.4 Ga match those reported from basement units in the hypothesized source areas for metasedimentary rocks in the EDC, which also overlaps with those from the WDC in this craton (e.g., Maibam et al., 2011, 2016; Figure 6a). Since the ancient sediments in the EDC occur as highly strained metamorphosed amphibolite-granulite facies rocks, primarily as thin inclusions and tectonic intercalations in terranes dominated by granitoid and orthogneisses, it is difficult to interpret the age data in the regional context vis-à-vis the WDC. The spread of minimum ages of detrital zircons from the metasedimentary rocks is quite large (2.7-3.4 Ga; Supplementary Table 3). Varied age ranges of studied EDC detrital zircons reflect (1) the provenance of the sediment, (2) temporal changes in the source of the sediments, (3) limited sampling. A minimum age for the onset of deposition in this block may be inferred from the presence of 2.9 Ga zircons in granitoids and charnockites that formed as a result of associated metamorphism (Mojzsis et al. 2003). Nonetheless the ages of studied detrital zircons and the reported older magmatic ages from the EDC establish the presence of a time of crustal growth at ~3.3 Ga (Figure 6a) prior to the voluminous emplacement of younger granitoids at 2.5-2.7 Ga (Figure 6c).

Studies of well-defined cores and rims in zircons from the metasedimentary rocks show the presence of 3.0 Ga overgrowths on 3.3 Ga or older cores. Although it is difficult to correlate the ages with specific thermal events, it appears that the ~3.0 Ga event may be tied up with the initial stabilization of the Dharwar craton, a major accretion event, (Hansen, Newton, Janardhan & Lindenberg, 1995) as well as a metamorphic episode at about 3.0 Ga (Figure 6B: Mahabaleshwar et al., 1995; Mahabaleshwar & Peucat, 1988; Peucat, Vidal, Bernard-Griffiths & Condie, 1989). We note that 3.0 and 3.25 Ga zircons have the same $^{176}Hf/^{177}Hf$ ratio, but the 3.0 Ga zircons have a negative εHf values. However, the sub-chondritic Hf-isotopic data suggest that the event represents reworking of pre-existing crust in line with the reported EDC granitoid intrusion and reported granulite metamorphism at 2.6–2.5 Ga (Jayananda et al., 2012, 2013). Combining our earlier reported data (Maibam et al., 2011, 2016) with the present dataset we infer that the geological processes in the EDC caused crystallization of new zircon with ages of 2.5–2.6 Ga as discrete grains (#32, Supplementary Table 3b, Supplementary Figure 1-xvi) and also as rims around older cores (#23, Supplementary Table 3b, Supplementary Figure 1-xi). The younger ages of overgrowths may be correlated with Dharwar volcanism in the greenstone belts and the late

Archean cratonization event marked by large-scale granite emplacement (Figure 6c). Zircons enclosed in garnets were also analysed, and their ages would refer to the age of metamorphism experienced by the rocks. Although the zircon ages from the thin sections are limited, the imprints of subsequent metamorphic events are recorded at 2.64–2.54, 2.74 and 3.0 Ga in the samples analysed in this study. The probable metamorphic ages could be deciphered from the sub-chondritic Hf-isotope ratios and low Th/U ratios of the conventionally processed zircons (Figure 4c) and are correlated to the reported metamorphic ages from the adjoining areas.

Zircon domains that yielded identical Pb-Pb ages show a more pronounced scatter of their initial $^{176}Hf/^{177}Hf$ (Figure 4d). It is important to emphasize that this scatter does not result from post zircon growth alteration, as the analysed zircons yield concordant U-Pb ages and the more inert Hf-isotopic system is still intact. Studies show that during high-grade metamorphic conditions $^{176}Hf/^{177}Hf$ ratios of magmatic and metamorphic zircon domains maintain the original ratios (e.g., Zeh et al., 2007; Gerdes & Zeh, 2009). This suggests that zircon crystallized over a short time interval from a melt, which clearly had a heterogeneous (not equilibrated) hafnium isotope composition.

Studies have shown that metamorphic zircon formed by a recrystallisation process will probably preserve its primary Hf isotopic composition even if the original U–Th–Pb information and zoning is obliterated, whereas metamorphic zircon that precipitates from a fluid or melt can have a distinct Hf isotopic composition because of incorporation of isotopically dissimilar Hf from external Hf reservoirs with differing Lu/Hf ratio (Gerdes and Zeh, 2009; Lenting, Geisler, Gerdes, Koojiman, Scherer, & Zeh, 2010; Patchett, 1983; Taylor, Kirkland, & Clark, 2016). Sláma et al. (2007) have shown that $^{176}Hf/^{177}Hf$ and Lu/Hf ratio of zircon crystals that grew at different stages of metamorphism during attendant, mineral destruction and growth processes, can potentially be used as tracers of zircon-forming metamorphic reactions in high grade rocks. Similar isotopic variations of the $^{176}Lu/^{177}Hf$ and $^{176}Hf/^{177}Hf$ ratios of the studied zircons of the ages are recorded (Figure 4a and 4d). The variation in the $^{176}Hf/^{177}Hf$ and $^{176}Lu/^{177}Hf$ ratios may reflect a mixture of the 'local' breakdown of Zr and Hf reservoir phases, such as garnet, rutile and Fe–Ti oxides (Bea & Montero, 1999; Beckman, Möller, Söderlund, Corfu, Pallon, & Chamberlain, 2014). Since rutile and Fe–Ti oxides occur as inclusions in the studied zircons, they represent plausible sources for Zr and Hf.

The combined U–Pb and Lu–Hf systematics of the zircons suggest the involvement of older crust (Paleoarchaean-late Eoarchaean) in the formation of Dharwar metapelitic rocks, no doubt due to erosion and reworking of these ancient granitoid during subaerial exposure (Figure 6a). In the εHf(t) vs. age plot (Figure 4b) the older zircon grains show chondritic to sub-chondritic values. The positive values of εHf suggest existence of depleted mantle domains formed in response to the primordial crust (Harrison, Blichert-Toft, Müller, Albarede, Holden & Mojzsis, 2005; Santosh, Yang, Shaji, Mohan, Tsunogae & Satyanarayanan, 2016). A signature of recycled crust is observed and the analysed grains also exhibit systematically increasing evolved isotopic signatures with time. Their distribution is mainly defined by an Eoarchean isotopic evolution line (Figure 4b), thus confirming the role of older recycled crust for the evolution of these rocks.

Our data suggest that the EDC gneisses were derived from recycled Paleoarchaean crust (cf. Maibam et al. 2016). Naha et al. (1993) suggested that older charnockite enclaves of amphibolite-facies gneiss occurred within the granulite-amphibolite transition area; however, their interpretations were not supported by geochronological or isotopic data. TTG gneisses with crystallisation ages ranging between 2.8-2.7 Ga and 2.5 Ga are reported (Hokada et al. 2013). The emplacement of the Closepet granite is associated with the migmatisation of Peninsular gneiss and the intrusion of discrete granite bodies nearly synchronous with charnockite alteration without resetting the older zircon isotopic characters (Mojzsis et al. 2003). Sample ED3 contains zircon ages ranging between 3.4-2.9 Ga and 2.7-2.5 Ga. These ages are coeval to the early crustal formation and reported major formation in the EDC (Mahabaleshwar and Peucat, 1988; Li et al., 2018). Widespread Neoarchaean (2.56-2.51 Ga) juvenile magmatism, associated with crustal reworking could have been responsible for the regional metamorphism (e.g., Peucat, Mahabaleswar & Jayananda, 1993; Li et al. 2018).

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FIGURE CAPTIONS

Fig. 1. Geological map of part of the Dharwar craton (modified from Harris & Jayaram, 1982) showing the outcrop locations from which samples ED3, ED6 and ED8 were collected.

Fig. 2. Outcrop photographs and thin-section photomicrographs of petrological features in the studied samples. Penknife is 15 cm long, and white scale bar in each photomicrograph is 1 mm. (A) Field photograph of metapelite sample ED6 showing stromatic leucosomes. (B) Large subhedral to anhedral orthopyroxene porphyroblasts in sample ED3, a high-Mg metapelite. These grains commonly contain globular quartz inclusions and are wrapped by weakly aligned biotite laths. (C) Metapelite sample ED6 contains texturally equilibrated plagioclase in leucosomes with ~120° angles at triple junctions. Minor myrmekite also occurs. (D) Garnet porphyroblast in sample ED8, a metagreywacke, with globular quartz inclusions and films of crystallized melt at porphyroblast margins (red dashed box).

Fig. 3. U–Pb probability density histogram showing the most likely ages obtained from LA-ICP-MS analyses.

Fig. 4. Results of combined zircon U–Pb dates and Lu–Hf isotope data (A) Lu–Hf (B) εHf (C) Th/U (D) $^{176}Hf/^{177}Hf$ and apparent Pb–Pb ages for all analyzed zircon grains (only analyses with degrees of concordance between 90% and 105% are considered in the plot). In Figure B, crustal evolution trend of average crust ($^{176}Lu/^{177}Hf$ = 0.0113) and mafic crust ($^{176}Lu/^{177}Hf$ = 0.022, Rudnick & Gao, 2003). Depleted mantle evolution trends DM1 and DM2, assuming a linear evolution of mantle depletion back-calculated from present day MORB (DM1 = 0.283165, Chauvel et al., 2008; DM2 = 0.283250, Chauvel & Blichert-Toft, 2001) using a mean $^{176}Lu/^{177}Hf$ of 0.0387. Note that DM1 trend would mean that the mantle depletion due to to crust formation has

not started before 4 Ga (see Zeh & Gerdes, 2012 for discussion) while DM2 trend assumes it started about 4.5 Ga ago.

Fig. 5. *P–T* pseudosections calculated for (A) ED3, a high-Mg metapelite; (B) ED6, a metapelite; and (C) ED8, a metagreywacke. Red dashed lines represent the calculated proportions of melt in each lithology. (D) Summary diagram showing the *P–T* conditions at which each observed phase assemblage would have been stable. White dashed arrow represents a possible retrograde evolution, based on the intersection of peak assemblage fields in each rock type with their respective solidi.

Fig. 6. Interpreted geological history of the studied metasediments from the Eastern Dharwar craton (EDC). Orange star represents the evolutionary path of the studied samples. (A) The proto-EDC crustal terrane experienced growth due to mantle plume-driven volcanism and intrusion, with erosion and weathering transporting material to intercontinental basins at c. 3.3 Ga. (B) Burial and crustal thickening at c. 3 Ga led to prograde metamorphism and zircon growth in the studied samples. (C) Final terrane accretion between the Eastern and Western Dharwar terranes led to further burial, metamorphism, and magmatism.

SUPPLEMENTARY MATERIALS

Supplementary Table 1. Representative EPMA-derived mineral compositions from all samples utilized in this study, shown in weight % oxides and cations per formula unit (cpfu). Proportions of Fe_2O_3 and Fe^{3+} were calculated using AX (Holland, 2009).

Supplementary Table 2. XRF-derived, whole-rock bulk compositions (weight % oxide) for samples ED3, ED6 and ED8. All iron is reported as Fe_2O_3 (i.e., Fe_2O_3(t)). LOI = loss on ignition.

Supplementary Table 3A. U–Pb isotope data for separated zircon from metapelite sample ED6.

Supplementary Table 3B. U–Pb isotope data for zircon grains separated from sample ED8 and included in garnet prophyroblasts.

Supplementary Table 4. LA-MC-ICPMS Lu–Hf isotope data of zircon from metapelite sample ED6.

Supplementary Table 5. Bulk compositions utilized for phase diagram modelling, given in mol. % oxide. FeO^{total} represents all iron as Fe^{2+}, where bulk-rock $X$$Fe^{3+}$ (= Fe_2O_3/(FeO + Fe_2O_3)) can be calculated as (2 × O)/FeO^{total}.

Supplementary Figure 1. Cathodoluminescence (CL; top) and backscattered electron (BSE; bottom) images of representative zircons grains in the studied samples. Circles (unbroken: U–Pb, dotted: Lu–Hf) analysed spots. Numbers: spot numbers and apparent Pb–Pb age.

Supplementary Figure 2. U–Pb concordia diagrams for zircon analyses from metapelite samples (A) ED6 (B) ED8 (C) Grains included in garnet porphyroblasts.