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REVIEW PAPER

Heavy minerals in provenance studies: an overview

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Abstract
Over the past few decades, the advent of sophisticated imaging and in situ measurements as well as data deconvolution techniques have led to remarkable progress in the field of heavy mineral research. The prevalence of zircon in a wide range of igneous, sedimentary and metamorphic rocks has been used frequently in estimating provenance, depositional age, tectonic settings, drainage evolution and crustal evolution. However, the biased age spectra (induced by hydrodynamic fractionation, sampling and measurement protocol and inheritance) yielded by detrital zircons reinvigorated the need to utilise other heavy mineral phases (monazite, apatite, titanite and rutile) for addressing a range of geological processes. Different heavy minerals are moderate to highly durable and provide variable response to magmatic and metamorphic events thereby providing clues that may be missed by single detrital grain analysis, thus emphasising the multi-mineral detrital approach as an indispensable method to investigate several geological processes. The present review highlights the role of detrital zircon and the associated limitations in using a single heavy mineral approach in geological studies. This review further emphasises the advantages of using multi-mineral/proxy studies and discusses the scope of heavy mineral research.

Keywords Detrital geochronology · Zircon · Provenance · Heavy minerals · Multi-mineral

Introduction

Provenance refers to the composition, location and source of the sediment/sedimentary rock (Schwab 2003). Provenance identification is vital in paleogeographic reconstructions, drainage evolution and characterisation of crust that seldom exists due to weathering (Haughton et al. 1991). Geochemistry of rocks/sediments and mineral extracted from them have been extensively employed to resolve the tectonics, magmatism, crustal evolution and provenance estimation (Morton and Hallsworth 1999; Lin et al. 2014; Joshi et al. 2017; Glorie et al. 2020; O'Sullivan et al. 2020; Armstrong-Altrin et al. 2021; Joshi et al. 2021a, b). Zircon is an extensively used mineral due to its wide distribution in igneous, metamorphic and sedimentary rocks, high closure temperature, resistance towards weathering, dissolution and diagenesis (Lee et al. 1997; Cherniak and Watson 2003; Ireland and Williams 2003; Malusà et al. 2013; Schoene 2013). Zircon also incorporates a vast number of minor and trace elements in its crystal structure along with substantial chemical and isotopic information (Finch and Hanchar 2003; Belousova et al. 2006; Kemp et al. 2007). In the last two decades, extensive efforts and rapid developments have been made in detrital geochronology (Joshi et al. 2021a).

The introduction of sensitive high-resolution ion microprobe (SHRIMP)/secondary ion mass spectrometry (SIMS) and Concordia diagram in geoscience stimulated the extensive attempts on uranium-lead (U–Pb) dating using zircon for the geochronology (Wetherill 1956; Andersen and Hinthorne 1972). The ion probe could analyse different growth zones within a zircon, while the Concordia made it convenient to appreciate the generated U–Pb dataset. To date, there have been considerable improvements in the mass resolution capacity of the ion probe. Additionally, the simultaneous advancements in imaging techniques [cathodoluminescence (CL), backscattered electron (BSE) and scanning electron microscope (SEM)] assisted in identifying various characteristics in a grain (magmatic and metamorphic) such as overgrowths, inclusions, fractures, microtextures and U content in each of the growth domains, thereby strengthening U–Pb zircon

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dating making it possible to investigate the transport medium and environment of deposition (Finzel 2017; Linnemann et al. 2018; Armstrong-Altrin et al. 2021).

In the 1980s, the ability of the Lu-Hf system as a potential geochronometer and tracer was demonstrated. Since then, a combination of U–Pb and Hf isotopes has been carried out on magmatic, metamorphic and detrital as well as extra-terrestrial zircons (Kemp et al. 2007; Iizuka et al. 2015; Sreenivas et al. 2019). The technological developments facilitated the measurement of oxygen isotopes in zircons by laser heating/gas-source mass spectrometry and ion microprobe/secondary ion mass spectrometer (Valley et al. 1994; Peck et al. 2001; Whitehouse and Nemchin 2009). The use of oxygen isotope in zircons has provided insights into the magmatic evolution of rocks, mantle geochemistry, provenance estimation and crustal evolution (Valley 2003; Hawkesworth and Kemp 2006; Bershaw et al. 2012; Hou et al. 2017).

Over the past few decades, a number of techniques have been adopted to estimate the age and other geological processes from zircons. The isotopic dating methods have been widely applied with a prime focus on detrital accessory minerals to ascertain age and associated geological processes. Detrital geochronology has an advantage over bedrock geochronology as it offers substantial coverage of regions that are inaccessible for sampling, weathered, or overlooked (Finzel 2019). Every accessory mineral used in dating yields different age distribution patterns which in turn suffices clues on the complex evolution of the source area (Vermeesch et al. 2009). In this contribution, we have attempted to review the role of detrital zircons in order to address: (a) its applicability in deciphering the provenance of sediments/sedimentary rocks; and (b) the benefits of multi-mineral/multi-proxy detrital approach over single grain detrital geochronology in comprehending the provenance.

Heavy minerals in geochronology

Zircon is a frequently used mineral for U–Th–Pb geochronology due to its robust nature and also because it incorporates U and Th with a trivial amount of Pb (Finch and Hanchar 2003; Ireland and Williams 2003). Zircon being resistant to chemical and mechanical weathering can archive other geological processes such as re-melting, variable cycles of sedimentation and sediment depositional history (Ireland and Williams 2003; Košler and Sylvester 2003). Studies from Jack Hills, Australia, have demonstrated that detrital zircons preserve ages (~4.40 Ga) much older than the oldest (~4.03 Ga) exposed bedrock lithologies (Bowring and Williams 1999; Wilde et al. 2001; Reimink et al. 2016). The U–Pb ages from zircon provide clues on crystallisation ages while hafnium and oxygen isotopes provide crucial information on mantle extraction time and sedimentary versus magmatic origin of zircons (Valley et al. 2005; Kemp et al. 2007; Iizuka et al. 2017). Therefore, detrital zircons have been extensively used to understand various geological processes like crustal evolution (Hawkesworth et al. 2010; Spencer 2020), provenance (Hietpas et al. 2011b; Sun et al. 2018), orogenic and tectonothermal events (Park et al. 2010; Sorcar et al. 2020), tectonic settings (Cawood et al. 2012; Shao et al. 2020), maximum depositional age (Dickinson and Gehrels 2009; Sharman and Malkowski 2020), paleogeographic and paleoclimate reconstruction (Cawood and Nemchin 2001; Dickinson and Gehrels 2008; Pullen et al. 2011; Zhao et al. 2020), paleo-drainage (Hurtig et al. 2020), denudation, exhumation and thermal histories (Sircombe and Freeman 1999; Welke et al. 2016; Chai et al. 2020).

Apart from zircon, the applicability of other accessory minerals such as apatite, monazite, rutile and titanite in age estimation have been realised with the continuous innovative attempts and improvements of the LA-ICPMS, SIMS and data reduction techniques. These advancements have led to precise U–Pb age and trace element estimation in various accessory minerals which have variable closure temperatures [zircon: >900°C (Cherniak and Watson 2001); monazite: 650–720°C (Suzuki et al. 1994); titanite: 550–650°C (Cherniak 1993); apatite: 450–550°C (Chamberlain and Bowring 2001); rutile: 400–500 (Mezger et al. 1989)]. These heavy minerals have different responses to magmatic and metamorphic events which aid in addressing numerous geological questions such as provenance tectonic settings, host rock compositions and metamorphic and cooling history of an orogen (Chew et al. 2011; Liu et al. 2014; Bruand et al. 2020; Pereira et al. 2020).

U–Pb, Hf and O isotopes from detrital zircon

The bulk rock/sediments trace and rare earth elements have been widely used to distinguish different source areas (McLennan et al. 1993; Basu et al. 2016; McLennan 2019; Banerji et al. 2021; Joshi et al. 2021b). However, numerous studies have underscored the proficiency of detrital heavy minerals (zircon, apatite, monazite, titanite, etc.) in addressing the rock/sediment provenance (Fedo et al. 2003; Gherels et al. 2011; Wu et al. 2017; O'Sullivan et al. 2020; Morag et al. 2021). Detrital heavy minerals especially zircons are widely applicable in identifying the source of sediments and sedimentary rocks (Košler et al. 2002; Lee et al. 2017; Armstrong-Altrin 2020; Armstrong-Altrin et al. 2021; Kong et al. 2021; Meinhold et al. 2021). Unlike petrography and geochemical studies, the investigation on detrital zircons for fingerprinting provenance needs limited information on the geology and geochemistry of the source regions (Vermeesch and Garzanti 2015). The zircon-based provenance identification involves the comparison of U–Pb ages obtained from detrital zircon of the studied site with the age spectra of the potential source regions (Howard et al. 2009; Armstrong-Altrin et al. 2017).

Mineral inclusions (feldspar, apatite, monazite, xenotime, muscovite etc.) armoured within zircons have also been utilised for source rock studies (Rasmussen et al. 2011; Bruand et al. 2016; Slabunov et al. 2017; Bell et al. 2018). A study based on the abundance of apatite inclusions from Jack Hills, Australia, suggested that the zircons were derived from felsic sources (Bell et al. 2018). Apatite is sensitive to change in magma composition and its trace elements can provide information on the whole-rock chemistry of the host rock (Jennings et al. 2011). A study based on the trace elements of apatite inclusions within zircons from late Caledonian plutons, Scotland, estimated the whole-rock SiO_2 and Sr content (Bruand et al. 2016). The study also invoked a possible linkage of U–Pb ages from host zircon and trace element abundances in the included apatite with the crystallisation history of the host phase, and thus, the information can be crucial in providing clues on the geochemical characteristics of the host magma.

The application of laser ablation multi-collector inductively coupled plasma mass spectrometer (LA-MC-ICPMS) and ion probe (SHRIMP/SIMS) had led to a revolutionary development in the provenance identification as they allow instantaneous measurement of trace elements and U–Pb/Hf isotopes on multiple zircon grains compared to TIMS (Ireland and Williams 2003; Košler and Sylvester 2003; Kylander-clark 2017; Xie et al. 2018; Joshi et al. 2021a). Further, the use of CL and BSE imaging techniques in conjunction with microanalytical instruments (LA-ICPMS, SIMS, etc.) escalated the study on unequivocal provenance identification. The BSE and CL images aid in deciphering various growth zones within zircon grains that can be individually analysed for age estimation and provide information on crystallisation as well as metamorphic history (Aleinikoff et al. 2006; Gehrels 2012; Zhong et al. 2018; Sorcar et al. 2020). This facilitates the evaluation of magmatic and metamorphic events (Davis et al. 2003; Thomas 2011), which provide clues on the source of the detrital fractions.

Generally, a few hundred detrital zircon grains from a sample need to be analysed for U–Pb and Hf isotopes to assertively demarcate the source of the sediments and sedimentary rock (Košler et al. 2002; Fedo et al. 2003; Hietpas et al. 2011a; Gehrels 2012). However, Thomas (2011) suggested that the obtained detrital age population should be treated with caution and the age spectra should be used in conjunction with the sediment dispersal paths for better control on provenance. The study suggested that the sediment dispersal paths may change with time due to shift in drainage pattern causing a shift in the source of the sediments. A recent study based on the variable ages obtained from the age spectra of more than 1300 detrital zircons from seven Eocene-Miocene sandstones and one modern Rio Grande River sand samples from south Texas suggested that there had been multiple shifts in the drainage pattern of the Rio Grande River (Fan et al. 2019).

In the past decade, it has been recognised that utilising only U–Pb ages in zircon for provenance identification can be spurious in regions influenced by detritus from multiple sources of similar ages (Howard et al. 2009). Using additional isotopic proxies like Hf and O isotopes from detrital zircons can place further constraints in provenance estimation. The Hf and oxygen isotopes when combined with U–Pb ages can be used to distinguish between the zircons derived from melting of igneous crust and those derived from melting of continental crust which contains a sedimentary component (Kemp et al. 2006; Howard et al. 2009; Dhuime et al. 2011). A few studies conducted from different stratigraphic units in Taiwan using U–Pb/Hf in detrital zircons and chemical ages from detrital monazite indicated variable sources for the sediments wherein the U–Pb and Hf isotopes in zircons suggested southeast China and Yangtze region as a dominant source, while detrital monazite age distributions invoked Yangtze block as a probable source (Yokoyama et al. 2007; Lan et al. 2016). The reassessment of the sedimentary provenance was conducted based on the U–Pb age spectra from detrital zircons of Lanyang River and Zhuoshui River in the east and west Taiwan, which inferred that the Eocene and Early Oligocene sediments were derived from Cathaysia Block, while Miocene and Pleistocene sediments were sourced from Yangtze Block and North China Block (Deng et al. 2017a, 2017b) (Fig. 1). This study substantiated the significance of detrital zircon from rivers draining major stratigraphic units as compared to those separated from sedimentary rocks.

The advantages of detrital zircon studies from modern river sediments were further corroborated by recent work on detrital zircon population from the Murchison River channel and Ordovician fluvial sediments (sandstones) in Western Australia that have drained similar sources (Markwitz et al. 2020). The U–Pb age distribution (Fig. 2), U content and 3-D shape of detrital zircons indicated a significant variation in detrital zircon population from sandstones and modern river sands. Further, the prominent underrepresentation of Archean ages in the sandstones compared to the detrital zircon age distribution from the modern river sands was also observed (Markwitz et al. 2020). Their study inferred that detrital zircon grains in modern river channels are stronger representative of their eroding source rock region than those contained within ancient fluvial sediments (sandstones).

The provenance estimation based on U–Pb and Hf isotope attempted on detrital and magmatic zircons extracted from para- and orthogneiss of Greater Himalayan Sequence (GHS) from Garhwal, north-west India, yielded an age range of 481–2560 Ma with εHf values ranging from −21 to 13 (Fig. 3) (Spencer et al. 2012). The reported U–Pb age range for paragneiss of GHS was in agreement with the previously reported values ranging from ca. 770 Ma to ca. 2600 Ma (Webb

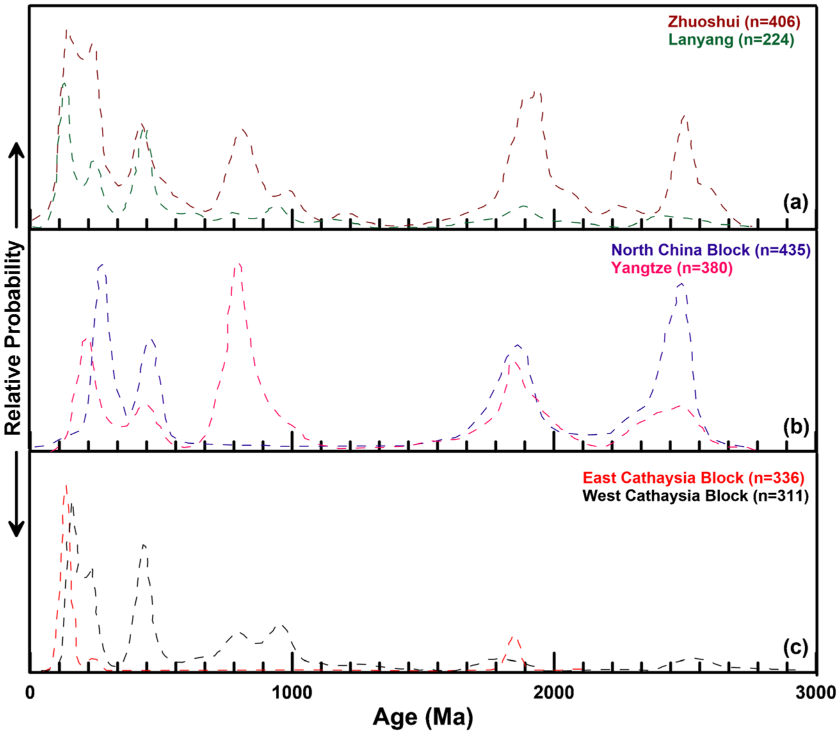

Fig. 1 KDE plots of U–Pb detrital zircons ages from Lanyang and Zhuoshui Rivers, Taiwan and Cathaysia, Yangtze and North China Block (Deng et al. 2017b)

et al. 2011) from SatlujRiver, Himachal Himalayas (Fig. 3), while the U–Pb ages for the orthogneiss ranged between ca. 416 Ma and 2740 Ma. Further, the younger zircon rims (~480 Ma) of the orthogneiss had εHf values from −11 to −7, while the εHf for the zircon core of orthogneiss (~860 Ma) ranged from −21 to 13 and were comparable with paragneiss (Fig. 3). The wide variation in the εHf values (Spencer et al. 2012) compared to those reported from the previous study (εHf from -24.5 to 5.2; (Richards et al. 2005)) from the Sutlej river valley revealed that the paragneiss was derived primarily from an 860 Ma continental arc, while the orthogneiss was sourced from the paragneiss of the Garhwal Himalayas (Spencer et al. 2012). Similarly, in the study from Gawler Craton, Southern Australia, the U–Pb ages (Archean to Early Proterozoic) of detrital zircons from paragneiss suggested derivation of the source sediments from the older portion of the adjacent Gawler terrane (Howard et al. 2009). However, Hf isotope for the same detrital zircons provided a strong signature of its derivation from the Pine Creek Orogen in the north Australian craton which was in agreement with the Nd bulk isotopic values (Howard et al. 2009).

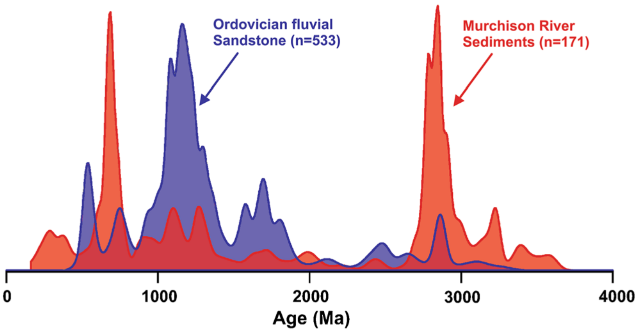

Fig. 2 KDE plots of U–Pb detrital zircons ages of Murchison River and Ordovician fluvial sediments, West Australia, showing significant variation in detrital zircon population from sandstones and modern river sediments (Markwitz et al. 2020)

Oxygen isotopes in combination with U–Pb ages and Hf isotopes can act as an additional proxy and help in delineating provenance with similar U–Pb age spectra. Hou et al. (2017) utilised the combination of radiogenic (U–Pb and Hf) and stable (Oxygen) isotopes from detrital zircons to decipher the source of clastic rocks interbedded with bauxite deposits in southwest China. Based on the whole-rock geochemistry, it was debated that the bauxite was sourced either from limestones or from flood basalts (Dai et al. 2007; Deng et al. 2010). The U–Pb detrital zircon ages (~260 Ma) obtained from clasts associated with bauxite suggested derivation from volcanic rocks (Hou et al. 2017). However, when these ages were seen in combination with εHf(t) (−26.6 to −0.6) and enriched $\delta^{18}O$ (+5.6 to +10.3 ‰) signatures, it was established that the sediments for bauxite deposit were sourced from felsic volcanic rocks from the volcanic arc at the northern margin of Paleo-Tethys (Hou et al. 2017).

The above studies reaffirm the fact that a single isotopic indicator from detrital zircons can yield erroneous or limited interpretations especially in the areas with multiple source contributors. Therefore, it is suggested that single isotopic proxies from detrital zircons should be used with caution.

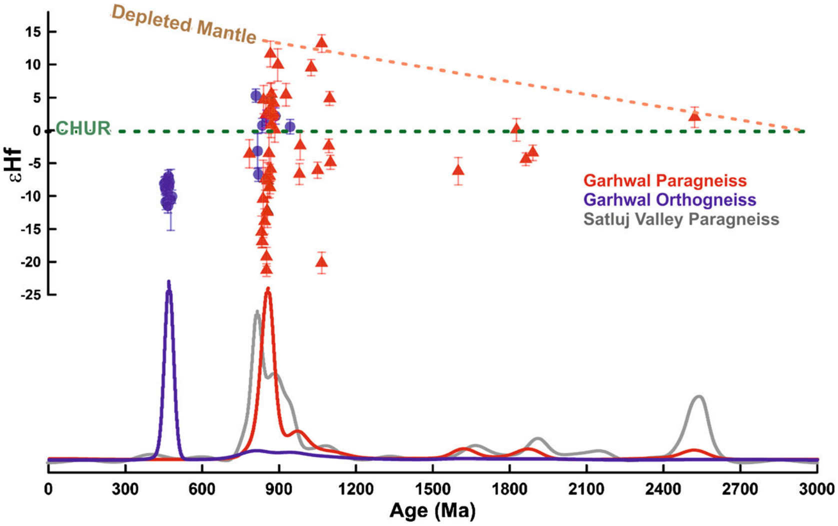

Fig. 3 U–Pb (KDE) and epsilon Hf values for the ortho and paragneiss from the Greater Himalayan Sequence, Garhwal, Himalaya, northwest India (Spencer et al. 2012). Comparative data for Satluj valley paragneiss (AW 9-22-04 10B) from Webb et al. (Webb et al. 2011)

These necessitate the implementation of multiple isotopic proxies from detrital zircons (U–Pb/Hf and oxygen) to interpret and identify the relevant source region.

Limitations of single mineral approach

Considering the resistant nature of zircons, the radiogenic and stable isotopes from detrital zircons have been widely used for crustal evolutionary as well as provenance estimation studies. However, single heavy mineral-based inferences can be affected due to biases induced by (i) hydrodynamic fractionation; (ii) sampling, sample processing and measurement protocol; and (iii) inheritance (Sircombe and Stern 2002; Moecher and Samson 2006; Hietpas et al. 2010; Sláma and Košler 2012; Malusà et al. 2016).

Hydrodynamic fractionation

Extensive statistics and precise instrumental errors are generally considered while inferring the detrital zircon values; however, the amount of hydrodynamic fractionation remains unconsidered (Malusà et al. 2016; Spencer et al. 2018). Such fractionation depends on various factors like depositional environment, the grain size of the host rock, zircon fertility of the bedrock, the erosion rate of the bedrock, catchment topography and degree of sorting. All these aspects can lead to spurious inferences based on the reported zircon population (Cawood et al. 2003; Moecher and Samson 2006; Malusà et al. 2016; Spencer et al. 2018).

The size of the zircons varies in igneous and metamorphic rocks and may depend on the grain size and zircon fertility of the host lithology (Corfu et al. 2003; Hawkesworth et al. 2019). The fertility of zircon in variable source rocks can amplify age signals from zircon-rich lithologies leading to skewness in the age spectra. A study conducted by Moecher and Samson (2006) on the granitoids from Carolina and Grenville basement terranes from North America suggested that the fertility of zircons can vary even within similar granitoid types. Their study also demonstrated that a higher modal zircon concentration in Grenvillian granitoids can enhance the number of detrital zircons compared to the granitoids from Carolina. It was further determined that the detrital zircon populations from both regions were significantly different. The difference was essentially due to high zircon content in Greenville terrane compared with low zircon content and its smaller size fraction from Carolina metavolcanic rocks (Moecher and Samson 2006). This study confirmed that the fertility of host lithology is of prime importance especially when the detritus contribution is associated with the inputs from mafic lithologies.

The detrital zircons travel a substantial distance and witness sedimentary recycling, which can lead to variable sizes that yield different ages. The ages obtained from such detrital fractions depend on the source and transportation history of the zircon (Lawrence et al. 2011; Garzanti 2016). Garzanti et al. (2018) suggested that the mineralogical composition of the sediments is a key factor that affects the grain size. Detrital heavy minerals are smaller in size and heavier as compared to quartz and feldspar minerals and, therefore, they tend to be concentrated in the finer sediment fractions (Garzanti et al. 2008, 2009). This sorting can skew the detrital age results (Lawrence et al. 2011; Ibañez-Mejia et al. 2018) and should be considered prior to age-dependent interpretation for addressing the provenance or crustal evolution. Based on the detailed analysis conducted on detrital fractions from the Amazon River basin, it was suggested that the U–Pb age from the zircon is dependent on the grain size wherein older grains are smaller on an average as compared to larger grains (Lawrence et al. 2011). However, later studies conducted to assess the relationship between size and U–Pb ages of detrital zircons revealed a minimal effect of grain size and hydraulic sorting on the age spectra (Garzanti et al. 2018; Markwitz and Kirkland 2018). A comparison of the detrital zircon age spectra from Nile delta beach and beach placer sands was conducted, and it was noted that the beach placer that had undergone extreme sorting had age spectra similar to that of delta beach sediments (Garzanti et al. 2018). Furthermore, their study compared the zircon age distribution with its size from Blue Nile sand, Ethiopia/Sudan and no clear dependence of grain size with age was invoked. This study further proposed an absence of grain size and hydraulic sorting effect on the detrital zircon age spectra.

Detrital zircon fractions of variable ages may behave differently during transportation and diagenesis. The older U-rich zircons may have undergone radiation damage and can be vulnerable to leaching and destruction (physical and mechanical) and lead to underrepresentation of age as compared to younger zircon fractions (Garzanti et al. 2009, 2018). This is further corroborated with the study conducted by Markwitz et al. (2020) who compared the detrital zircon population from Murchison River in Western Australia with Ordovician sediments (Fig. 2). They concluded that the Archean zircons were underrepresented in Ordovician sediments and suggested that crystal fragmentation of metamict zircons were responsible for such skewed results.

Sampling, processing and measurement

The sample separation techniques (crushing and sieving) and analytical methods adopted for the measurement of isotopes can also result in U–Pb age bias (Sircombe and Stern 2002; Fedo et al. 2003; Moecher and Samson 2006; Sláma and Košler 2012). The heavy minerals are generally separated either through heavy liquid separation or using a magnetic separator. The study conducted on detrital fractions of fuchsite quartzite from Slave craton, Canada, suggested that the age bias was related to paramagnetism, which was restricted to metamict zircons of Archean age (Sircombe and Stern 2002). Post magnetic separation, the minerals of interest are either handpicked randomly or based on colour and size for further analysis. Experimental results from synthetic detrital fractions revealed that the zircon population represented by small zircons grains was underrepresented in age spectra (Sláma and Košler 2012). This was typically due to the preferential handpicking (for the ease of picking and mounting) of larger zircon grains. The sampling and separation technique may have a negligible effect on zircon ages for sediments from a single provenance, but it can induce substantial age bias if the sample consists of multiple provenance inputs with variable grain size (Sircombe and Stern 2002; Sláma and Košler 2012).

Further, in most cases, the images of the grain zoning are attained before analysis and the core zone is specifically selected for ablation as it is large enough for 30–40-μm beam chosen for the analysis (Moecher and Samson 2006). Under such circumstances, the rim of the zoned mineral grain remains unanalysed due to its smaller size (not sufficient for chosen spot size) and thus can further augment the age bias during such measurements. Further, measurement protocols, calibration standards, fractionation corrections and choice of using ^{207}Pb/^{206}Pb for older to ^{206}Pb/^{238}U for younger ages can also lead to measurement age bias (Nemchin and Cawood 2005; Sláma and Košler 2012). The approach in plotting U–Pb values has also recently raised concerns as in most cases, the probability density plots (PDPs) are utilised for density estimation from detrital U–Pb zircon values which remains theoretically unexplained (Vermeesch 2012). Another statistically reliable alternative method called kernel density estimator was proposed for plotting detrital age distributions (Vermeesch 2012). Thus, the choice of using PDP or Kernel Density Estimate (KDE) may also cause bias in the estimated age peaks from U–Pb zircon values. Different groups using different protocols from a similar detrital population can therefore add significant variability to the age spectrum.

The data generations for zircon ages using U–Pb isotopes were initially limited (due to the intensive process involved in the TIMS measurement) but the use of LA-ICPMS and SIMS has led to U–Pb analysis in hundreds of detrital grains (Pullen et al. 2014). The sophisticated imaging techniques (CL and BSE imaging) further helped in overcoming the grain scale sampling pitfalls arising due to complex zoning in zircons, until sampling can cause bias in the results (Hietpas et al. 2010). A few hundred analyses from a single sample are often used to decipher the source or provenance of the sediments while the bias caused by fractionation, sorting, sample selection and/or processing remains unnoticed (Sircombe and Stern 2002; Lawrence et al. 2011; Pullen et al. 2014). Such interpretations or sampling may not be representative and can be unsuccessful in retrieving the entire range of age populations leading to skewed results. A large number of analysis on detrital grains can significantly aid in identifying low abundance age components unless age bias is induced during sample preparation (Pullen et al. 2014). The required number of analyses depends on the proportion of variable ages present in the sample, inheritance and the precision of the analytical method used (Gehrels 2012). However, a recent study on detrital zircons from Rio River, Venezuela, revealed that even a large number (n=1000) of analysis from a sample is insufficient to adequately characterise the complete spectrum of detrital zircon ages available for a fluvial system (Ibañez-Mejia et al. 2018). A similar study conducted for provenance estimation in French Broad River of the north Carolina and Tennessee emphasised the possibility of substantial variability in the age spectrum of the detrital zircons despite the samples being collected within a few kilometres, while incomplete or biased source signatures may be recorded if only limited samples were collected for detrital studies (Fig. 4(a)) (Hietpas et al.

2011b). This was further confirmed by the estimated U–Pb age for nearly five samples collected within the same micro-environment (single location) of the Amazon River which attested the presence of a statistically variable age spectrum from all the analysed samples (Lawrence et al. 2011).

The above studies underscored the natural and artificially induced limitations of single sample analysis in capturing the entire provenance signatures. Thus, in order to get geologically robust information from the detrital zircon ages, multiple samples from the same system should be analysed.

Inheritance

Limited zircon growth in marginally magmatic thermotectonic events paves additional shortcomings in the age estimation and provenance studies. Zircon formation is commonly found in convergent plate boundaries while it is rarely present in low-temperature melts, low-grade metamorphic and major tectonic events due to strike-slip plate motions (Hietpas et al. 2010; Piechocka et al. 2017). Zircons from such terranes will miss collisional, crustal thickening, heating and loading, melt generation and exhumation events and will only record inheritance, and the analysed ages will certainly lead to biased ages with skewed age spectrum towards the older source (Moecher and Samson 2006; Hietpas et al. 2010; Be'eri-Shlevin et al. 2018). The impact of the selected heavy mineral in provenance estimation was reflected by the investigation conducted on detrital monazites and zircons from the sandstone of the Appalachian Foreland basin (Hietpas et al. 2011a). The study revealed that the zircon age spectra from Lee and Pocahontas Formations failed to record the younger (Taconian and Acadian) Appalachian events which were well documented in detrital monazite spectra. The observed skewness of age spectra (towards older ages) from detrital zircons of the six sandstone samples underscores the limitations of using zircons for provenance estimation in localities dominated by metamorphic rocks (Fig. 4(b)). This robust nature of zircon obscures precise provenance estimation.

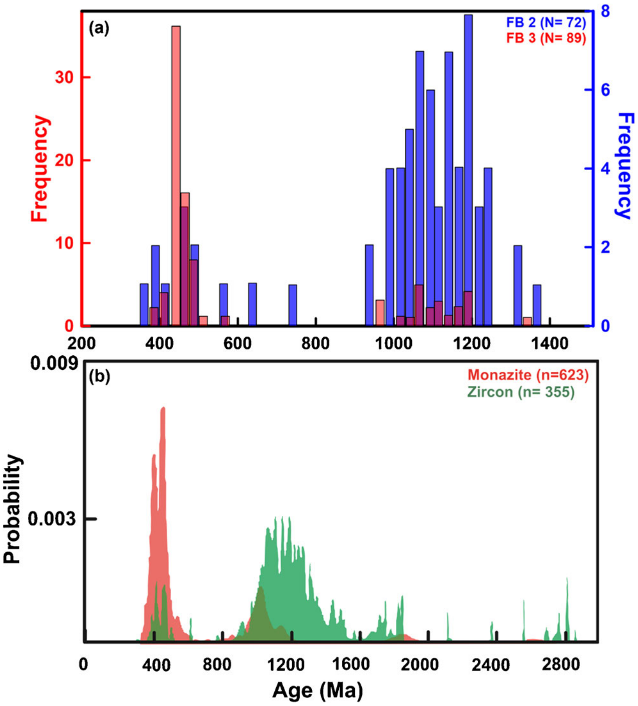

Fig. 4 (a) Histogram showing variation in U–Pb detrital zircon age spectra for two selected samples of French Broad River collected a few kilometres apart (Hietpas et al. 2011b). (b) Detrital zircon and monazite age spectra from Appalachian Foreland basin showing the impact of the chosen heavy mineral in provenance studies (Hietpas et al. 2011a)

Multi-proxy approach in provenance estimation

The limitation of the single mineral approach has been emphasised in the previous section wherein the detrital zircons can yield biased ages or might miscue certain events or lithologies with recessive zircon abundance. This lacuna can be catered by using a multi-mineral proxy approach to understand various geological processes. Due to their variable closure temperatures, the heavy minerals (zircon, rutile, monazite and titanite) provide different responses to magmatic and metamorphic events thereby providing clues that may be missed by single detrital grain analysis (O'Sullivan et al. 2016; Kirkland et al. 2017; Guo et al. 2020; Joshi et al. 2021a; Liu et al. 2021).

Multi-mineral studies

Dating of detrital zircons by SIMS or LA-ICPMS has in many ways acted as a valuable tool in deciphering sediments provenance. Some studies suggested that the detrital zircons can precisely constrain the source terranes, provided the protosources have not been masked by recycling and mixing of material from different sources with similar age ranges. The recycling of the detrital zircon via successive erosion-transportation and mixing may homogenise the material from different sources thereby limiting the zircon as a source indicator (Hietpas et al. 2011a, 2011b; Zack et al. 2011; Andersen et al. 2016). Under certain instances, the zircon-based provenance studies may not be able to fully identify all the source terrains (Hietpas et al. 2011a). Therefore, the trace element and isotope systematics from detrital apatite, muscovite, rutile and their combination have been successfully implemented for provenance studies in the past decade (Be'eri-Shlevin et al. 2018; Malusà et al. 2017; O'Sullivan et al. 2018; Sun et al. 2018). Depending on the heavy mineral and the proxy used, each mineral/proxy offers distinct insights on the source (Guo et al. 2020).

Zircon having high closure temperature has limited growth below upper amphibolite-granulite facies conditions (Kohn et al. 2015). This makes it unresponsive towards low- to medium-grade metamorphism and tectonic events (O'Sullivan et al. 2016). The utility of detrital monazite and zircons for provenance indicator was attempted for the French Broad River, Appalachian Blue Ridge and Carboniferous–Permian sandstones from the Appalachian foreland basin (Hietpas et al. 2010, 2011a). These studies suggested skewed age spectra towards Mesoproterozoic or older ages indicated by the detrital zircon due to its robust and refractory nature and, thus, failed to record the multiple collision events from the Appalachian orogen (Fig. 4(b)). Similar observations of preserved older ages (dominance of Grenvillian zircons) were suggested for the detrital zircons from Modern Appalachian Rivers (Eriksson et al. 2003). However, detrital monazite successfully recorded the complex Paleozoic orogenic events which were missed by detrital zircons (Fig. 4(b)) (Hietpas et al. 2010, 2011a). As monazite is less resistant than zircon during diagenesis, hence it is less likely to be recycled as compared to zircon (Eriksson et al. 2003; Hietpas et al. 2011a). Additionally, monazite crystallises over a wide range of metamorphic conditions than zircon, thereby providing the evidence on moderate thermal metamorphism that were missed by detrital zircon (Rubatto et al. 2006; Högdahl et al. 2012). The results ratified the applicability of monazites in deciphering Appalachian orogeny/regional metamorphism (~450–470 Ma) and the inheritance dominant age spectra from detrital zircons. However, monazite can also induce bias towards metapelites lithologies (O'Sullivan et al. 2016). Thus, the application of detrital apatite and rutile on collisional mountain belts was investigated from the French Broad River, Southern Appalachians (O'Sullivan et al. 2016). In these studies, the detrital rutile and apatite recorded Appalachian metamorphic event at ca. 320 Ma which was associated with the assembly of Pangea (Fig. 5). However, this event was missed by detrital monazite (ca~450–470 Ma) by a previous study (Moecher et al. 2011). In view of this, O'Sullivan et al. (2016) suggested that apatite and rutile can provide cues on medium-grade metamorphic events due to their lower closure temperatures. A similar approach of multi-mineral analysis for deciphering provenance and evolution has been applied in recent studies from Lhasa, Nianchu and Pumchu rivers, Tibet (Guo et al. 2020), and Merrimack River, New England, USA (Gaschnig 2019). Based on the detrital multi-mineral (zircon, monazite, titanite and rutile) approach, the studies revealed variable age and source response for different heavy minerals and reaffirmed the applicability of multiple heavy minerals in seeking provenance and crustal evolutionary histories of complex orogens (Gaschnig 2019; Guo et al. 2020).

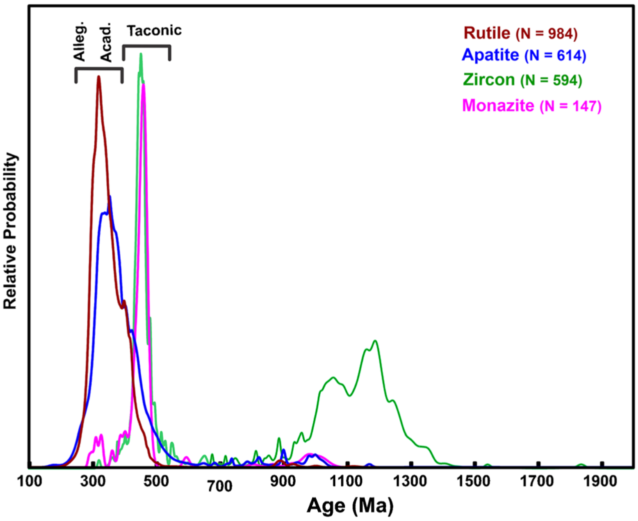

Fig. 5 KDE plots of U-Pb ages of detrital rutile, apatite, zircon and monazite from French Broad River demonstrating the applicability of various detrital minerals for the investigation of provenance studies in complex metamorphic terrains (O'Sullivan et al. 2016 and references therein). Alleg., Alleghanian; Acad., Acadian

Multi-mineral or multi-proxy studies

Recently, a number of studies have employed different combinations of multiple proxies like Raman spectroscopy, microstructures, U–Pb geochronology, radiogenic and stable isotopes and trace element chemistry of heavy minerals, and sediment chemistry for provenance estimation (Armstrong-Altrin et al. 2018; Gillespie et al. 2018; Ansberque et al. 2019; Ramos-Vázquez and Armstrong-Altrin 2019; Resentini et al. 2020; Armstrong-Altrin et al. 2021). A recent study exploited surface microtexture, mineralogy, bulk sediment geochemistry, trace element composition and U–Pb isotopic geochronology of detrital zircons from Riachuelos and Palma Sola beach areas, southwestern Gulf of Mexico, to decipher the provenance of the sediments (Armstrong-Altrin 2020). On the basis of morphology, microtextures (collision fractures, V-shaped percussion cracks, abraded grains with bulbous edges, angular grains with broken edges), key trace element ratios (Th/U >1) of zircon and major (low SiO_2) and trace element (elevated Sc, V, Co and Ni, enriched LREEs, negative to positive Eu anomaly) sediment chemistry (Fig. 6(a)) suggested that the majority of the sediments for both Riachuelos and Palma Sola beach were sourced from felsic to intermediate igneous rocks. Consequently, they were transported by littoral and aeolian currents which were deposited in a high-energy subaqueous environment. Further, the dominance of Paleozoic and Cenozoic zircons (Fig. 6(b)) indicated that the Zacatekas and Nazas Formation in Mesa central province and Eastern Alkaline Province were

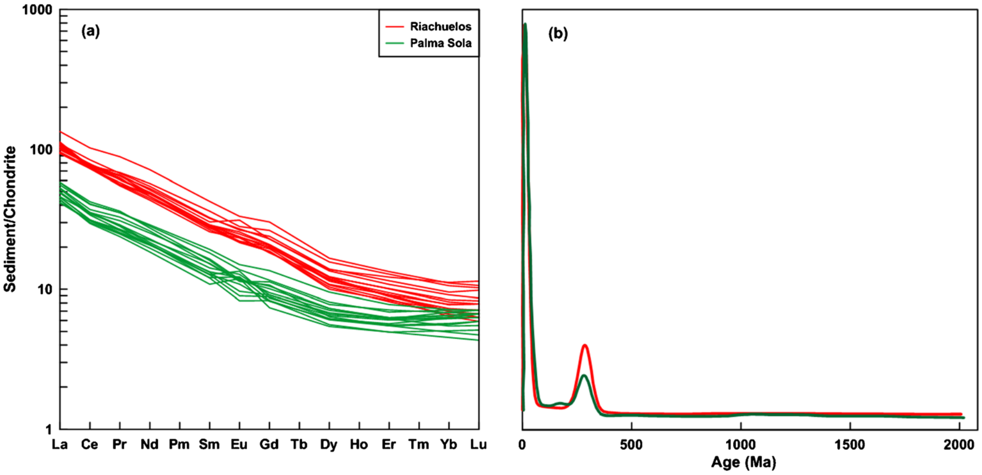

Fig. 6 Chondrite normalised rare earth element distribution patterns for the Riachuelos and Palma Sola beach sediments. Chondrite normalisation values are from Sun and McDonough (Sun and McDonough 1989). Geochemical and U–Pb isotope data for Riachuelos and Palma Sola beach sediments (Armstrong-Altrin 2020)

the dominant contributors. The presence of limited Proterozoic zircons made them propose a possible contribution from the Grenvillian igneous suites in the Oaxaca and the Chiapas Massif Complexes. The lack of correlation between U–Pb zircon ages and rare earth elements from Riachuelos and Palma Sola beach sediments invoked that U–Pb age spectra is a preferable approach for provenance discrimination as compared to zircon rare earth elements (Armstrong-Altrin 2020). However, a similar study on Atasta and Alvarado beach sands, western Gulf of Mexico, advocated that variation in the zircon rare earth element patterns can also be beneficial in delineating the variation in sources (Armstrong-Altrin et al. 2018).

In order to understand the crustal growth histories and provenance, several studies have utilised ^{40}Ar/^{39}Ar ages in detrital mica/hornblende collectively with double dating by using U–Pb and He in (U–Th/He) detrital zircon (ZHe) and (U–Th–Sm)/He apatite (AHe) (Welke et al. 2016; Sun et al. 2018; Zhuang et al. 2018; Zotto et al. 2020). Detrital zircon U–Pb ages and ^{40}Ar/^{39}Ar ages from mica can be successfully used in provenance studies, wherein robust zircons record the magmatic and high-grade metamorphic histories while ^{40}Ar/^{39}Ar ages in mica record more recent tectonic history (McDougall and Harrison 1988; Harley et al. 2007; Harrison et al. 2009; Kohn et al. 2015). On the other hand, ZHe and AHe ages obtained from detrital zircons and apatite can be utilised to differentiate similar U–Pb ages by identifying the cooling ages (Reiners et al. 2004; Welke et al. 2016). The double dating of detrital zircons from feldspathic arenite and quartz arenite from the central Appalachian Basin presented ZHe ages of 300–475 Ma which were much younger than the detrital zircon ages (ca. 1000 Ma) (Zotto et al. 2020). In this study, the observed time lag between U–Pb detrital zircons and ZHe ages suggested that the sediments had undergone multiple recycling episode post exhumation. Another study from Hindu Kush-Kohistan-Karakoram utilised U–Pb detrital zircon geochronology and ^{40}Ar/^{39}Ar thermochronology on detrital mica from rivers, and they identified pre- and post-India Asia collision events and synchronous cooling events (Zhuang et al. 2018).

The information from radiogenic and non-radiogenic isotopes from detrital zircon can be further complemented with Nd isotope measurements in detrital apatite, monazite and titanite which act as an important repository and potentially control the majority of the rare earth element budget (Bea 1996; Tiepolo et al. 2002). The Sm–Nd systematics from different accessory minerals with variable preservation potential can record melting events and changes in magmatic sources. Therefore, it is widely applied in dating metamorphic events and for deciphering crustal evolutionary studies in terranes with complicated thermal histories (Hammerli et al. 2014; Zhou et al. 2020). Multi-mineral and multi-proxy study was carried out on zircon and monazite from modern rivers draining South China Block for crustal evolutionary studies (Liu et al. 2017). Their data also depicted skewed zircon ages spectra towards older ages; however, a considerable overlap was noted in the Hf (zircon) and Nd (monazite) model ages obtained from both the minerals. Their study concluded that Nd isotopes from monazite, like Hf isotopes in zircons, can track the crustal evolution and growth of the continental crust. They further suggested that studies from modern river sediments should utilise both detrital zircon and monazite to develop a better picture of the evolutionary record.

An integrated multi-mineral approach utilising U–Pb/Hf/Nd isotopes in detrital zircons, apatite, monazite, rutile and titanite grains was undertaken on the sediments from Yong-Ding and Luan Rivers to comprehend the tectonomagmatic evolution of the study area (Zhou et al. 2020). Both these rivers drain the catchment areas of the northern North China Craton (NCC) and variable portions of the southern Central Asian Orogenic Belt (CAOB). Their study demonstrated that detrital zircons with U–Pb ages in the range of 2.3–2.6 Ga, 1.8–2.0 Ga and 0.13–0.38 Ga from the river sediments archived the magmatic episodes in the region, while two

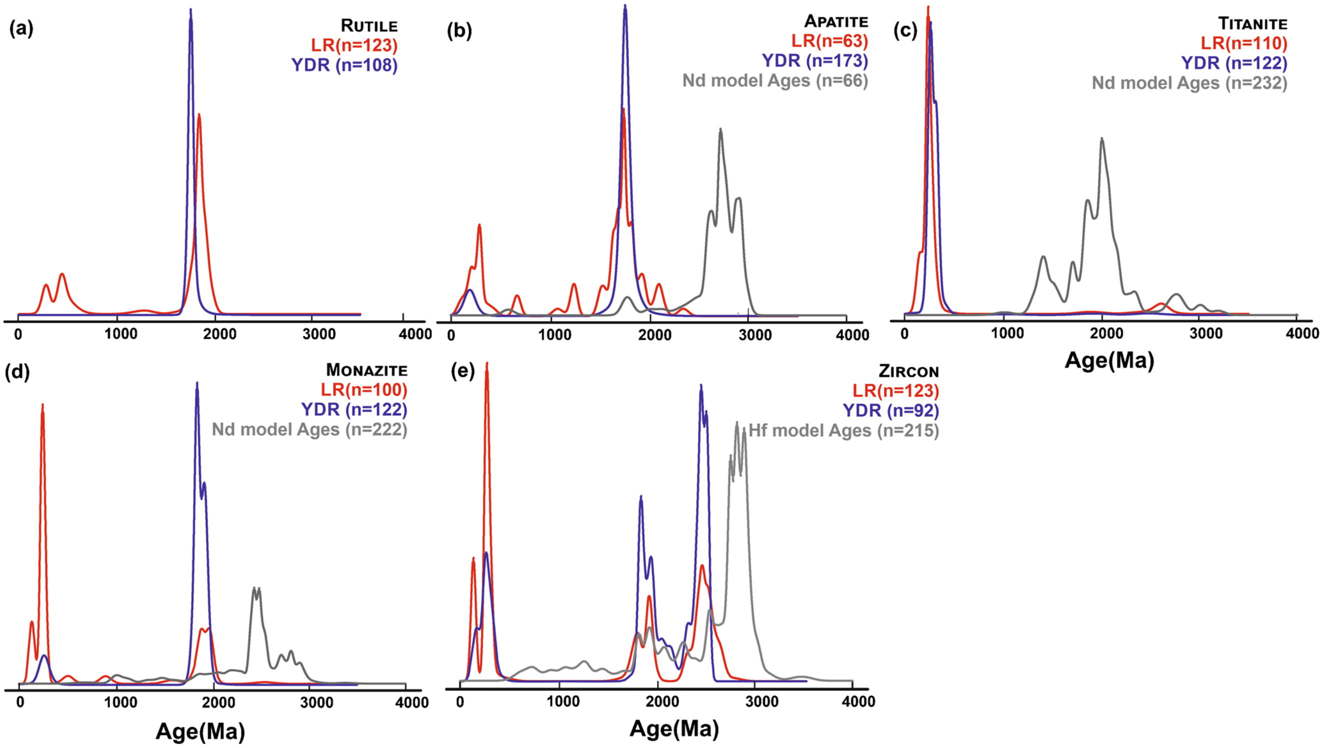

Fig. 7 (a–e) KDE U-Pb, Hf and Nd model age spectra for the Yong-Ding River (YDR) and the Luan River (LR) revealing the advantages of multi-mineral and multi-isotopic approach in the complex orogen (Zhou et al. 2020)

metamorphic events at around ca. 1.85–1.95 Ga and ca. 1.6–1.9 Ga were recorded by the U–Pb of monazite (Fig. 7). On the other hand, ages yielded by apatite and rutile provided the cooling ages following the Proterozoic collision. Further, it was concluded that the detrital ages from titanite were biased towards Phanerozoic calc-alkaline magmatism and were unsuccessful in recording the older metamorphic or magmatic imprints. The Hf and Nd model ages from zircon, monazite and apatite suggested a significant crustal growth from 2.7 to 2.9 Ga which was well documented from NCC (Fig. 7). However, it was noted that Hf isotopes in zircons were unsuccessful in recording post-Archean crustal evolution of the region which was documented by monazite at ca. 2.5 Ga and ca. 1.2 Ga and titanite at 1.8–2.2 Ga and 1.3–1.5 Ga (Zhou et al. 2020).

The above studies demonstrate that it might be inappropriate to infer the past geological events based on a single heavy mineral and isotope, especially in a complex orogen. Therefore, a multi-mineral and multi-proxy approach is recommended wherein the resolving power of various detrital phases can be combined with various proxies to assess the complex geologic history of the terrane. Since, different accessory minerals have their own capabilities and limitations, and thus, it is suggested to adopt a multi-mineral approach to precisely decipher the provenance and petrogenetic evolution of complex terrane (Sun et al. 2018; Jiao et al. 2020).

Summary and scope

In the past few decades, zircon dating has paved an essential foundation in providing cues on the source and validating various geological processes. However, the limitations such as hydrodynamic fractionation, inheritance, sampling, processing and data reduction techniques have led to biased or spurious age and provenance interpretations. Thus, it is important to shift from a single mineral to multi-mineral and multi-proxy approach for understanding source to sink relationships. The provenance and crustal evolution investigations have been found to be abstruse in complex orogenic terranes with the influence of multiple metamorphic events (O'Sullivan et al. 2016; Guo et al. 2020). Under such circumstances, the applicability of other detrital heavy minerals can act as additional chronometers and facilitate in reconstructing petrogenetic histories and source information (O'Sullivan et al. 2016; Bruand et al. 2017; Zhou et al. 2020).

Presently, the comparison of rare earth elements with isotopic signatures from different heavy minerals is seldomly implemented. The oxygen isotopes from zircon have been explored in crustal evolutionary research; however, oxygen isotope for other heavy minerals remains inadequate (Bindeman 2008; Bruand et al. 2019). The technical advances have reinvigorated the analyses of in situ O, Nd, Cl and H isotopes from titanite and apatite (King et al. 2001; Foster and Carter 2007; Bonamici et al. 2014; Tartèse et al. 2014; Bruand et al. 2019). Such isotopic parameters should be further combined with mineral inclusion studies from detrital accessory phases for demonstrating source composition, metamorphic histories, provenance, crustal evolution and magma discrimination (Bruand et al. 2014, 2020; Hart et al. 2016; Bell et al. 2018). The applicability of mineral inclusion should be verified on heavy minerals from modern river sediments for revealing their source rock compositions and petrogenesis.

In the Indian subcontinent, substantial U–Pb zircon ages exist for detrital zircons from ancient sediments but insufficient detrital zircon investigation in modern river sands has been attempted. Despite the usefulness of the various isotopic techniques, still there exists a lacuna in the heavy mineral research in the Indian subcontinent. Recently, a few studies have combined bulk rock chemistry, Sr–Nd isotopes and U–Pb ages of detrital monazites/rutile with the detrital zircons from clastic rocks to re-evaluate the provenance in western India and Eastern Ghats Granulite Belt (Mandal et al. 2019; Axelsson et al. 2020; Chaudhuri et al. 2020b). Nonetheless, there still exists a gap in utilising combined multiple heavy minerals (zircon, rutile, titanite, monazite, etc.) geochronological approach to address or refine provenance and crustal evolutions from ancient and modern sediments. Recently, the heavy minerals (garnet, rutile and tourmaline) chemistry from modern sediments (east coast) and sandstones (Kutch basin) of the Indian subcontinent highlighted that the relationship, as well as the ratios between key trace elements from the heavy minerals, can provide clues on source lithologies (Naidu et al. 2019; Chaudhuri et al. 2020a). Such a multi-mineral approach, when supported by U–Pb ages and other chemical and isotopic proxies, can act as an indispensable tool for provenance interpretations.

The future of heavy mineral studies lies in using multiple accessory minerals for U–Pb dating so that the resolving power of various detrital phases can be utilised to address provenance as well as petrogenetic histories of complex terranes. These studies should be combined with bulk rock chemistry and Sr–Nd isotopes as a preliminary approach. Ultimately, the need of the hour is to utilise the ocean of information generated from multi-mineral and multiple proxy approaches which will open a new window to interrogate the rock and sediment record through time and is the future of heavy mineral research in Earth sciences.

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Availability of data and material (data transparency) Present work does not include any data.

Code availability (software application or custom code) Present work does not utilise any code.

Declarations

Competing interests The authors declare that they have no competing interests.

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