Earth and Planetary Science Letters ••• (••••) ••••••

Contents lists available at ScienceDirect

Earth and Planetary Science Letters

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A zircon petrochronologic view on granitoids and continental evolution

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A R T I C L E I N F O

Article history:
Received 10 July 2019
Received in revised form 29 October 2019
Accepted 3 December 2019
Available online xxxx
Editor: R. Dasgupta

Keywords:
detrital zircon
geochronology
trace element chemistry
continental crust
crustal evolution

A B S T R A C T

Temporal trends in granitoid chemistry and thermometry constrain major global changes in magmatism, tectonism or crustal thickness in the continents. Our study relies on zircon geochronology and trace element geochemistry on four new detrital rocks (two modern sediments and two Archean metasedimentary rocks) and a global compilation of published single zircon detrital chronology and trace chemistry data acquired on 5587 individual grains. Zircons of all ages from 4.4 Ga to present exist in this archive. Ti-in-zircon thermometry indicates that more than 98% of the grains with concordant U-Pb ages formed at temperatures exceeding 650 °C. The great majority of these zircons formed in the 650–850 °C range consistent with growth in intermediate to silicic magmas. Magmatic temperatures increased over time for the first 1.2 Ga of Earth's history after which they stayed constant before decreasing during the more recent past. U/Th < 5 values in the overwhelming majority of grains are consistent with a magmatic origin. La/Yb, Sm/Yb and Eu/Eu* values are relatively constant throughout the history of the Earth suggesting that most granitoids formed at, or evolved from magmatic reservoirs located at depths of 35–45 km in the presence of amphibole, garnet and limited plagioclase. Such reservoirs are common today in hot deep crustal environments beneath some of the thicker island arcs and all continental arcs along subduction zones. Processes other than modern day style subduction may have contributed to the formation of granitoids in the early Earth but temperatures, depths and the presence of water arbitrated by the presence of amphibole were similar. These results also suggest that the thickness of continental crust in areas that produced granitoids was similar to today's global average throughout the 4.4 Ga time period covered by the zircon archive. There is no correlation between zircon chemistry over time and the assembly of supercontinents.

1. Introduction

A new era in understanding global geologic processes commenced about a decade ago with the arrival of large geologic, seismic, paleontological, geochemical and geochronological databases that summarize information otherwise too difficult to compile and digest by single investigators or groups (i.e. GEOROC, EarthChem, etc.). Continental tectonics and evolution, which has been a subject of great interest, speculation and controversy for over two centuries and has been at the center of new seminal papers that make use of these large and growing datasets (e.g. Keller and Schoene, 2012, Hawkesworth et al., 2016). It is clear that our geologic archive is an incomplete one marking the competing effects of formation, destruction and ultimate incomplete preservation of various products, igneous, sedimentary of metamorphic rocks, fossils, etc. The distribution of zircon U-Pb ages for example (Cawood et al., 2013) is inhomogeneous at a global scale, with striking peaks and lulls in age distribution, or at least in zircon abundance (Voice et al., 2011). That is somewhat puzzling given that the Earth must

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https://doi.org/10.1016/j.epsl.2019.116005
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maintain some form of steady state evolution in order to balance crustal formation and recycling (Arndt, 2013; Hawkesworth et al., 2016). It becomes therefore critical to be able to distinguish in these large databases global evolutionary patterns from various preservation biases (i.e. Cawood et al., 2013). This most likely will require several iterations of hypotheses among the Earth scientists of the day but beyond lies the genuine possibility of a new transformative global understanding of Earth's tectonics at its 4.5 billion years evolutionary timescale.

The research of continental crustal evolution has tapped into the ever-growing open-access global databases of igneous rock geochemistry since igneous processes provide the principal mechanisms for generating rock and chemical diversity over time in the Earth's crust. Lesser used but viable alternative materials believed to capture average chemical crustal compositions are fine grained sediments (such as glacial tills, e.g. Gaschnig et al., 2016). Recently, more ambitious attempts have been made using this array of whole rock geochemical data and forward models to unravel global compositional and, among other parameters, crustal thickness changes over the entire lifespan of the Earth (Dhuime et al., 2015). These global and long-term compilations and their interpretations are aimed at deciphering major changes, whether abrupt or gradual in the overall evolution of continental masses, as driven by tectonics (Hawkesworth et al., 2016) or some other major known changes in the Earth's evolution such as the great oxygenation event at the end of the Archean. It is becoming increasingly clear that crustal tectonics evolved over time, and several distinctive evolutionary stages can be defined (Hawkesworth et al., 2016); at this point these are put forward as working hypotheses. As such, as many types of data as possible need to be employed in order to check for internal consistency in this search for global trends in the evolution of continental masses.

Zircons preserved in the detrital archive span almost the entire range of Earth's age (Cawood et al., 2013) but in addition to age they provide critical information about the origin of the magmas from which they crystallize. Those include Hf and oxygen isotopes (e.g. Vervoort and Blichert-Toft, 1999, Valley et al., 2005), temperature of crystallization (Watson et al., 2006) as well as the lesser used concentrations of various trace elements (e.g. Nagasawa, 1970; Claiborne et al., 2010; Grimes et al., 2015). All of these are measurable today via *in-situ* laser ablation techniques, making the zircon archive crucial in deciphering continental evolution. Of the elements whose trace elements are measurable in zircon, the rare earth elements (REE) are perhaps the most useful in deciphering the origin of parent magmas using zircon-melt partition coefficients (Watson, 1980, Hoskin and Ireland, 2000; Whitehouse and Kamber, 2002; Luo and Ayers, 2009, Nardi et al., 2013).

Here we report new age and chemistry data on detrital zircons from a variety of young sediments draining large rivers as well as some old metasedimentary rocks. The goal is to capture regional changes in zircon chemistry. We also incorporate previously published data in an attempt to provide global evolutionary pathways for REEs in granitoid rocks of the continental crust. We show that there is little change in these parameters over time and most zircons measured in this study and previous ones grew in equilibrium with granitoids with geochemical characteristics similar to those found in Andean arcs today and at relatively constant depths of 35–45 km.

2. Zircon partition coefficients

The use of detrital minerals allows us to access similar information over large temporal and spatial scales. Partition coefficients for zircon and intermediate magmas are in general well established (e.g. Geochemical Earth Reference Model – GERM), although they vary significantly from study to study. In a recent study (Chapman et al., 2016), we used an empirical technique in which we determined whole rocks and zircon REEs from the Coast Mountains Batholith to determine general partition coefficients for zircon-intermediate melt. The zircon-intermediate melt partition coefficients for lanthanides are dependent on concentrations of the elements in zircon, which is an unexpected and poorly understood departure from Henry's law (Chapman et al., 2016). These partition coefficients predict whole rock concentrations of REE from various plutonic and volcanic rocks more accurately than those obtained in previous work (e.g. Fujimaki, 1986, Luo and Ayers, 2009, Nardi et al., 2013). We use the Chapman et al. (2016) partition coefficients here in order to calculate the concentrations of REEs in the whole rocks from which the zircons analyzed here crystallized. The same partition coefficients are applied to the literature data used in this study.

3. Samples and methods

We present new detrital zircon age and geochemistry data on two modern sediment samples (Danube sample has been collected from a heavy fraction enriched sand bar along the Danube delta and Yangtze sample was combined from two locations within the cities of Wuhan and Shanghai, China). In addition, we measured two ancient metasedimentary rocks, a meta-sandstone from Witwatersrand, South Africa and a metasandstone from Jack Hills, Australia. Sampling locations, coordinates and references are presented in *Supplementary appendix 2*. The Danube and Yangtze rivers were selected because they drain geologically complex regions that include young orogens as well as cratonic areas. On the other hand, in order to expand the timeline coverage up to Early Earth, the Witwatersrand and Jack Hills samples were selected. In addition to new data obtained from these samples, we complied a global database of previously published ages and trace element concentrations in zircon. This database may not be exhaustive, but it contains all the data available to these authors at the time of manuscript writing.

Sample preparation is described elsewhere (Gehrels et al., 2008). Analytical technique details for U-Pb age determination can also be found in Gehrels et al. (2008) and with up to date modifications presented in https://sites.google.com/a/laserchron.org/laserchron/. All SEM investigations, U-Pb isotopic analyses for zircons were performed at the University of Arizona in the Arizona Laserchon (ALC) facilities (Gehrels et al., 2008). U-Pb ages (based on isotopic ratios) and trace element concentrations in zircon were measured simultaneously on an Element 2 high resolution ICP-MS with an E2 excimer Photon Machine laser ablation system. We targeted larger than usual detrital populations, ideally around 300 zircon grains per sample although this was not possible in all cases. We report "best ages" depending on analytical uncertainties of the $^{238}U/^{206}Pb$ ages (more accurate for young, <1.4 Ga ages) and $^{207}Pb/^{206}Pb$ (more accurate for old ages). We rejected age values that have more than 10% discordance between isotopic clocks.

Trace and Rare Earth Elements in zircon provide a powerful tool for studying petrogenetic processes of igneous samples and for reconstructing provenance and source terrane characteristics for detrital samples. In Supplementary Appendix 1 we outline the methods that have been developed for analysis of trace and rare earth elements by LA-ICPMS at the Arizona LaserChron Center (ALC).

4. Results

Each sample population consisted of over 300 individual zircons, some of which were rejected. Data are reported in Supplementary Appendix 2. Rejection was primarily due to age discordance, and/or Ti-in-zircon temperatures outside of the realm of reasonable magmatic origin (<600 °C and >1300 °C). Prior to

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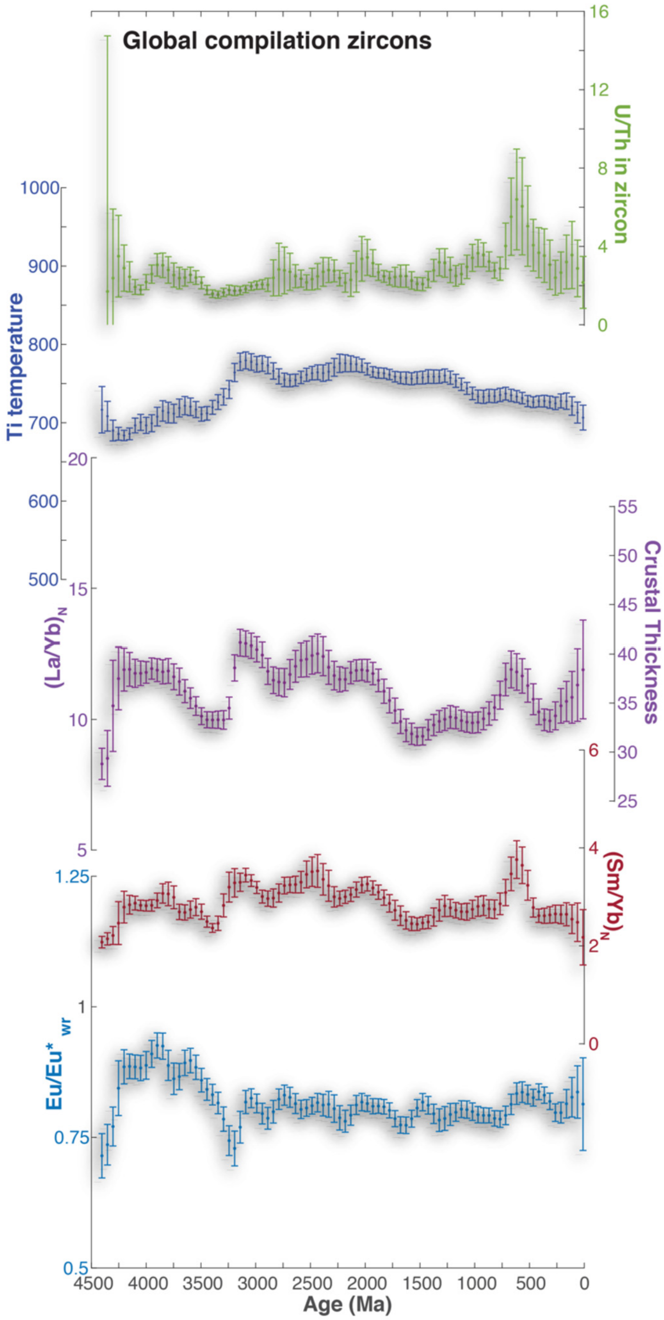

Fig. 1. Zircon variability U/Th and Ti temperature, and of calculated whole rock (wr) Eu anomaly, $(Sm/Yb)_n$, and $(La/Yb)_n$ throughout Earth's history. Data were produced using Monte Carlo and bootstrap resampling techniques (see appendix 4 for details) on a 5587 points dataset (previously reported added with 887 new data). Bootstrapped mean values are with 2σ uncertainties for age bins of 50 Ma.

eliminating data-points based on temperature, we did notice that low temperature zircons do not necessarily have high U/Th, which is normally linked to a metamorphic origin (e.g. Hoskin and Schaltegger, 2003). About 10% of the data gathered in this study were discarded due to the causes listed above. Sources of previously published age and chemistry data are summarized in the Supplementary Appendix 3. The combination of these samples with previously published data represent the data pool for interpretations in this paper, which in sum consist of 5,587 individual data points.

We present graphs for the aggregate global database (our new samples and literature data). Fig. 1 summarizes the global data pooled at 50 Ma intervals using the methodology detailed in Supplementary Appendix 4: Ti-in-zircon temperatures (Watson et al., 2006), U/Th ratios in zircon, La/Yb_n Sm/Yb_n ratios as well as Eu anomalies (Eu/Eu*, calculated as $Eu_n/\sqrt{Sm_nGd_n}$) in the whole rock equivalents. A few results stand out in our samples as well as the global dataset. Most zircons have U/Th <3 and are correlated with temperatures in the range of 600–900 °C, strongly indicating a magmatic origin for the great majority of the zircon archive (e.g. Hoskin and Schaltegger, 2003). Moreover, the range of temperatures recorded by the Ti-in-zircon thermometer on over 92% of the zircon population is similar for all geologic times, from the Archean to present. This range of temperature covers most granitoids, from the minimum eutectic melts of leucogranites (∼620 °C) to those typical of making intermediate magmas (tonalites and granodiorites) by dehydration melting of mafic protoliths (850–950 °C, e.g. Rapp and Watson, 1995). A distinctive increase from 700–800 °C pre-3.2 Ga followed by a gradual decrease since then is evident in the global database.

There are no correlations in our database between La/Yb and U/Th, nor is there any pattern of correlations between La/Yb or U/Th and the temperature of magmas (not pictured). There are large variations in these plots but no obvious correlations. U/Th does appear to correlate weakly with the magnitude of the Eu anomaly calculated for the whole rock values – this is evident in our samples as well as in the global compilation. The only strong correlation between geochemical parameters is within ratios of REE: La/Yb correlates well with Sm/Yb, as well as with the Eu/Eu*.

Calculated whole rock La/Yb_n are within a range of ratios (10–15) throughout the Earth's history without a clear pattern of decrease or increase over time – this is observed in individual samples (not pictured) as well as in regional global averages. The average La/Yb_n for today's arc rocks as obtained from whole rock values (Profeta et al., 2015) projects back into the past to the earlier Archean as calculated from the zircon archive. This would correspond to a crustal thickness of about 35–42 km if the Profeta correlation is used. To a first order, global zircon data shows a noteworthy constancy of calculated whole rock La/Yb_n from the Archean to present. However, notable spikes in La/Yb_n exist: there is a clear increase in the Hadean, another distinct and marked positive spike at the critical 3.3–3.1 Ga (see below), a more gradual decrease in the mid-Proterozoic, and finally a positive spike at the very end of the Neoproterozoic and early Cambrian (650–520 Ma), following the breakup of Gondwana (e.g. Cawood and Buchan, 2007). This latest spike is well correlated with the Sm/Yb_n ratios as well as an increase in U/Th, but not in temperatures and Eu/Eu*.

5. Discussion

5.1. Origin and temperatures of zircons

Over 92% of the zircons investigated here (new and previously published data) display Ti-based temperatures and U/Th ratios consistent with an igneous origin. To a first order, the archive of detrital zircons available in the continental crust preserves a record of igneous processes over time, and primarily that of making granitoids. While many zircons are found in metamorphic rocks, they are either pre-metamorphic and "opaque" to metamorphism even at high temperatures (Rubatto and Hermann, 2007) or for those who do form during regional metamorphism, they make up a minuscule fraction of the global detrital budget.

Moreover, the vast majority of zircons record temperatures in the range of 650–850 °C which is indicative of equilibration with granitoid melts. This is not surprising and consistent with experimental data on zircon saturation in magmas (Watson et al., 2006) as well as observational data (Miller et al., 2003) that intermediate rocks such as those formed in Phanerozoic subduction-related magmatic arcs, yield the most zircons of any magmatic rocks. Consequently, the zircon archive analyzed here is to a first order an interpretation of how intermediate rocks such tonalites, granodiorites or their more ancient equivalents, the tonalite-trondhjemite-granodiorite suites, formed over time. The detrital zircon cargo thus may be relevant to making the continental crust as a whole to the extent to which the average of the continental crust is that of a tonalite-granodiorite (Rudnick and Gao, 2013).

The global or regional databases presented here show a decrease in average zircon temperatures over time after about 3.2 Ga, suggesting that post Paleo-Archean zircon producing granitoids formed basically within the same temperature range but perhaps mimicking the overall cooling of the Earth. Earlier zircons appear to show a distinct increase in temperature from the earliest zircons of the Hadean towards the end of the Archean, from around 620 to 900 °C. This distinctive and apparently long-term increase in zircon temperature in the global database can have multiple interpretations but overall since it took place within the same general depth range (see below) it must mark the change from an eutectic melting of the Ab-An-Q system towards progressively higher dehydration melting of biotite and amphibole-bearing rocks.

5.2. *Trace element chemistry*

We discuss the slopes of the La/Yb_n and Sm/Yb_n as well as the Eu anomaly (Eu/Eu^*) calculated for the whole rocks in equilibrium with zircons. The first ratio can be related to crustal thickness calculations, or is at least an indicator of shallow (low La/Yb_n) *versus* deep (high La/Yb_n) fractionation, the second is a good marker of amphibole-dominated ($Sm/Yb_n < 4$) *versus* garnet-dominated ($Sm/Yb_n > 5$) fractionation of granitoids, whereas the Eu anomaly (numbers significantly below 1) indicates shallow processes and plagioclase present. To a first order, most granitoids formed with minor Eu/Eu^* and elevated La/Yb_n throughout all the investigated samples and the surveyed literature. If crustal thickness correlations were to be used, crustal thicknesses of about 35–45 km are calculated from most zircons and that correspond to La/Yb_n of around 10–20. Yangtze and Danube samples display fluctuations in La/Yb_n over time with a periodicity of 200–300 Ma indicative of Wilson cycles that are inevitably marked by periods of thinning alternating with periods of crustal thickening, but they are within the high La/Yb_n range of modern arcs. The Sm/Yb_n ratios are generally <4 suggesting that amphibole had a major role in fractionating the REEs, perhaps in the presence of garnet but certainly not dominated by garnet as in the case of eclogite melting. Eclogite melts have much steeper slopes of the MREE/HREEs as well as La/Yb (Martin et al., 2005; Castillo, 2012). Simply put, there is overwhelming evidence from these zircons that granitoids formed throughout the Earth's history were primarily derived from 30–50 km deep MASH-type zones (Annen et al., 2006) in the presence of amphibole and garnet ± plagioclase (Davidson, 2007, Lee and Anderson, 2015), similar to modern/young continental arcs such as the Andes or the western North American Cordillera (Ducea et al., 2015). Overall, this apparently critical depth of granitoid fractionation (35- to 45 km) is where granulitic residues give way to arclogitic ones (Lee and Anderson, 2015) in modern sub arc environments; that transition can span 20 km depth or more (Ducea and Saleeby, 1998). Our regional data (individual new data points) is marked by some fluctuations over time in these geochemical tracers, but they only cover a fraction of the variation seen in igneous rocks today at various tectonic settings. At global scale, the zircon geochemical database is remarkably uniform over time, leaving no room for interpretations favoring secular changes in the origin of granitoids.

5.3. *The 3.x Ga event*

Some lines of evidence suggest that at 3.3 to 3.1 Ga ago or thereabout (referred here to as "3.x Ga") a major change in crustal evolution occurred (Dhuime et al., 2015) and after a significant event of crustal recycling, much of the continental crust present today began to emerge. Our zircon compilation geochemical data suggest a major increase in La/Yb, Sm/Yb and decrease in Eu/Eu^* took place at around 3.3–3.1 Ga, a step of far greater magnitude than any other geochemical change in our dataset at any other time. The same interval sees a prominent increase in Ti temperatures bolstering the idea that a major change in crustal evolution took place at about that time. Our data do not directly constrain or rule out a process but strongly support the idea of a relatively sharp turn in continental evolution at 3.x Ga, which also seems to be the time recording the apex of zircon (and thus granitoid) temperatures globally.

5.4. *Implications for crustal thickness*

What do these results teach us about continental crustal thickness over time? They are certainly not suggesting that all continental crust was 35 ± 10 km thick from the Hadean on, but because zircon producing granitoids did form by either partial melting or fractionation of basaltic protoliths/melts (Lee and Anderson, 2015), this process was on average buffered at those depths. The presence of mafic materials at 40 km depths is an indication that either the crust was that thick in such environments or slab melting took place at those depths (Harrison, 2009). Relatively low temperatures and the indication that amphibole was present among the fractionating phases are similar to modern subduction settings. In fact, the continental crust today averages about 42 km (Mooney, 2007) and all parameters investigated here project toward similar numbers today as they were in the past.

There is ample evidence that today and the past billion years plate tectonics is different than plate tectonics of the early Earth (Harrison, 2009; Kemp et al., 2010; Hawkesworth et al., 2016) although some form of plate tectonics may have existed back in the Hadean and Archean (Harrison, 2009) and might not have operated in full extent (Cawood et al., 2018). Extensive oceanic (basaltic) shallow slab melting may have taken place early on. Alternatively, smaller scale plume and delamination (vertical) tectonics could have been responsible for basalt formation and ponding and subsequent evolution into granitoids at tens of kilometers beneath the surface.

5.5. *The Neoproterozoic spike*

A distinct positive peak in La/Yb_n and in Sm/Yb_n, which most likely translates into higher crustal thickness, occurs in the Neoproterozoic possibly extending into the Cambrian (650–520 Ma), coincident with the beginning of dispersion of Gondwana. This is a time broadly coincident with the Snowball Earth event, a time where most of Gondwana's continental mass was located near the South Pole. Neoproterozoic magmatism is known throughout the globe, with large batholiths forming notably in the vicinity of the Arabian Nubian shield (Stern, 1994) immediately prior to or during the collision between west and east Gondwana. A steep climb in Sr isotopic ratios culminating at around 500 Ma (Veizer et al., 1989, i.e. relatively soon, some 50–70 My after the crustal thickness maximum in our data) is also consistent with erosion of basement typically attributed to times when more than usual high standing masses exist. Oceanic and transitional arcs are well known in the latest Precambrian (Triantafyllou et al., 2018) but there is no immediate first order knowledge that the continents or at least the arcs formed at that time were thicker than normal. If that

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is the case, perhaps the unusual amount of erosion experienced by the continents at that time (Keller et al., 2019), which is attributed to Snowball glaciations, could also be driven by higher average elevations commonly accompanying thicker crust. Fig. 2 shows that overall this Gondwanan frame shows some of the most pronounced spikes in global databases (oxygen, Hf isotopes, but not in the Rb/Sr-driven crustal thickness). This potentially important temporary increase in global crustal thickness needs to be further tested using regional whole rock data from Gondwanan basement terrains containing igneous rocks that span that entire age range and beyond.

5.6. Other interpretations

We note that the relatively few Hadean zircons in the database are consistent with many previous studies suggesting that the crust was significantly thinner then (e.g. Harrison, 2009), which is an intuitive expectation as well. The drop in La/Yb_n ratios at the potentially critical time of about 3.6 Ga could be related to global changes in tectonic regime, from melting basalts from sub oceanic plateaus to more modern-like plate tectonics generation of granitoids (Bell et al., 2014). This has been observed in exposed Eo-Archean terrains via changes in Sr/Y, La/Yb_n and other chemical parameters in the classic TTG (tonalite-trondhjemite-granodiorite) assemblages. The gradual early Proterozoic decline of La/Yb_n in our data to a minimum during the Neoproterozoic may or may not have a significance in terms of marking the formation of more island arcs (with thinner crust).

We note that our La/Yb_n evolution path is to a first order not consistent with the Dhuime et al. (2015) Rb/Sr curve for crustal thickness (Fig. 2), which suggests a rather smooth increase of crustal thickness over time. These global databases will have to be iterated again as more data becomes available. There is also no obvious correlation between the global abundance of zircons of various ages and most of our chemical parameters (not pictured). It appears that abundance of zircon is to a first order correlated to super-continental cycles (Condie, 2014) and geochemical parameters investigated here are not, except for the Gondwana moment.

5.7. Implications for continental formation and tectonics

The classic hypothesis of continental crust formation via oceanic arc accretion (Taylor and McLennan, 1985) is somewhat at odds with the data presented here in that modern island arcs tend to form on thin crust and have distinctively lower in La/Yb_n and have larger negative Eu/Eu* (Profeta et al., 2015). Granitoids from which zircons grew, evolved from deep crustal magma chambers or remelting zones and, to the extent these zircons come from granitoids that average those of continental chemistry as a whole, they did not form in thin crust associated with Mariana-like subduction margins. Simply put, granitoids that make up the continents seem to have been produced at some similar depths throughout the Earth's history despite the different processes (plumes earlier, plate tectonics later) that may have produced them. True eclogite derived slab melts produce much larger La/Yb_n and no Eu anomalies, and they are unlikely to have been a significant player in granitic magmatism in the past just as they are not significant today. One important take home message from the zircon geochemistry database is that granitoids and probably the continental crust form by hot zone processes in "factories" located at 30–40 km beneath the surface regardless of which tectonic stage the Earth was living through. This interpretation also indirectly argues that some parts of the continental crust were that thick for much of the 4.4 Ga of granite making on the planet.

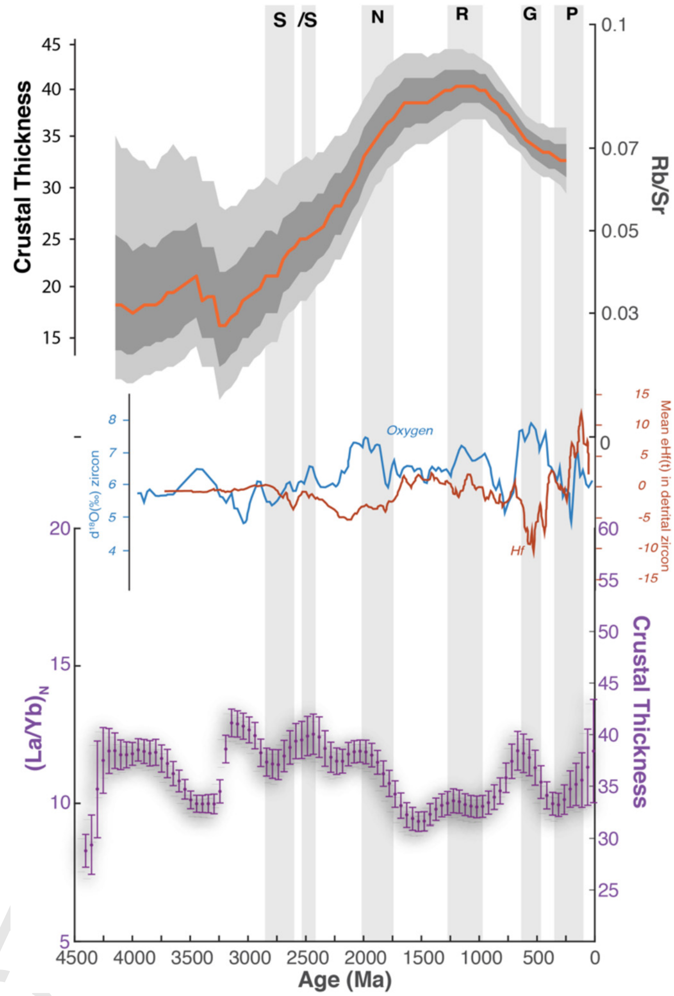

Fig. 2. Global whole rock $(La/Yb)_n$ variation along 4.5 Ga of Earth's history versus other major global landmarks in zirconology: Hf, oxygen isotopes (Hawkesworth et al., 2016), and Rb/Sr (Dhuime et al., 2015). Gray shades mark supercontinent assembly intervals (Cawood et al., 2013; S/S – Superia/Sclavia, N – Nuna, R – Rodinia, G – Gondwana, P – Pangaea).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

We acknowledge support from US National Science Foundation grant EAR 1725002 and the Romanian Executive Agency for Higher Education, Research, Development and Innovation Funding project PN-III-P4-ID-PCCF-2016-0014.

Appendix A. Supplementary material

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.epsl.2019.116005.

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XMLVIEW: extended

Appendix A. Supplementary material

The following is the Supplementary material related to this article.

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Label: MMC 1
caption: Analysis of U-Pb geochronology and trace elements in zircon at the Arizona LaserChron center.
link: **APPLICATION : mmc1**

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Label: MMC 2
caption: Location of samples analyzed in this paper and U-Pb and trace elemental concentrations of detrital zircons from each sample.
link: **APPLICATION : mmc2**

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Label: MMC 3
caption: U-Pb ages and trace elemental concentrations of literature data used in this paper. A separate worksheet provides a reference list for various sources used in this compilation.
link: **APPLICATION : mmc3**

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Label: MMC 4
caption: Methodology for pooling ages.
link: **APPLICATION : mmc4**

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Highlights

- A global survey of age – chemistry of zircons shows similar features of granitic magmatism over time.
- Changes in temperature and chemistry do exist over time in the global database.
- 3.2 Ga is a moment of great change in continental evolution.
- A Neoproterozoic spike in crustal thickness and metamorphism is notable in the database.

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