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Emergence of peraluminous crustal magmas and implications for the early Earth

M.R. Ackerson^{1*}, D. Trail², J. Buettner³

Abstract





Detrital zircons from the Jack Hills (JH) metasedimentary belt of Western Australia are a record of the first ~1.5 billion years of Earth history and can be used to help reconstruct the conditions of crust formation and secular changes therein. Beginning as early as *ca.* 4.3 Ga, but becoming more pronounced in the mid-Archean, a peraluminous signature begins to emerge from the JH zircon record. Combined with trace elements (P, REEs) and Ti-in-zircon thermometry, this increase in peraluminosity is likely the result of deep (>7 kbar) partial melting of hydrous mafic protoliths or partial melting of metasedimentary source material. In a geodynamic context, these results may suggest a gradual shift from a vertical tectonic regime toward a horizontal tectonic regime with potential subduction-like or collisional

processes creating the necessary conditions for peraluminous melt generation beginning locally at least by ~3.6 billion years ago (Ga).

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Introduction

Continental crust and its derivatives (*e.g.*, detrital zircons) preserve a near continuous record of Earth's history from ~4.4. Ga to today (Voice *et al.*, 2011) that can be used to investigate both individual magmatic systems (*e.g.*, Reimink *et al.*, 2014) and secular changes in Earth's geodynamic and tectonic history (Bauer *et al.*, 2020). The ~3 Ga metasedimentary rocks from the JH in Western Australia contain detrital zircons that are the oldest known record of Earth's continents (Compston and Pidgeon, 1986). These ancient zircons retain multiple chemical fingerprints that have been combined to reconstruct a Hadean Earth that bore oxidised, water-rich silicic continents derived from mafic (±felsic) protoliths that interacted with low temperature (T) surface waters (Cavosie *et al.*, 2005; Watson and Harrison, 2005; Trail *et al.*, 2011; Burnham and Berry, 2017).

One outstanding problem in Earth's early history is the nature and timing of the transition from a stagnant lid or vertical tectonic regime (crust thickened by processes akin to modern oceanic plateaus; Van Kranendonk *et al.*, 2004; Reimink *et al.*, 2014) to a mobile lid or horizontal tectonic regime where crust can be thickened and rapidly recycled through subduction-like processes and/or shallow thrusting (Bauer *et al.*, 2020). Horizontal tectonics is associated with an increase in the depth of melt generation. The shift toward a horizontal tectonic regime should produce chemical signatures in the ancient rock record that reflect this geodynamic shift. One potential tool to evaluate this shift is the Al content of zircon crystals (X^{Zrc}_{Al}, in ppm by weight), which is strongly dependent on the peraluminosity

of the melt from which the zircon crystallised (Wang and Trail, 2019).

Experiments and natural observations indicate that peraluminous melts (here defined as those with an aluminum saturation index [ASI = molar Al₂O₃/(CaO + Na₂O + K₂O)] > 1 (Shand, 1943) are the product of a limited number of petrogenetic processes. Here, we investigate X_{AI}^{Zrc} from the JH as an indicator of peraluminous melts and relate this to petrogenetic models in the context of geodynamic shifts in the first ~1.5 billion years of Earth's history.

Samples and Results

Zircons were collected from clastic metasedimentary units within the Jack Hills metamorphic belt (Weiss *et al.*, 2015). All zircons were separated from their host rock and mounted in epoxy using previously described techniques (Trail *et al.*, 2017; also see Supplementary Information). Care was taken to ensure the trace element compositions reported for zircons are primary structure bound signatures (*e.g.*, not metamict regions, mineral inclusions, secondary alterations). Data were discarded if: 1) Fe > 100 ppm, 2) analyses outside 100 ± 10 % concordance, and/or 3) light rare earth element index (LREE-I) < 50 (Bell *et al.*, 2019).

The cumulative zircon age distribution is similar to previously reported ages from the JH (*e.g.*, Crowley *et al.*, 2005), with notable age peaks at 3370 and 4050 Ma (Fig. 1). Exceptionally, site T145 contained a high proportion (15.5 %) of unaltered Hadean-aged zircons. Zircon titanium concentrations are similar

^{*} Corresponding author (email: ackersonm@si.edu)



¹ Department of Mineral Sciences, National Museum of Natural History, Smithsonian Institution, Washington, DC

² Department of Earth and Environmental Sciences, University of Rochester, Rochester, NY

³ Los Alamos National Laboratory, Los Alamos, NM



Figure 1 (a) 207 Pb/ 206 Pb JH zircon age distributions from multiple units. (b) Ti in JH zircons with (c) Ti-in-zircon temperatures, calculated assuming $a_{TIO_2} = 0.5$ and $a_{SIO_2} = 0.8$ (Ferry and Watson, 2007) with zircons from the LFB (Trail *et al.*, 2017).

to other studies of the JH (*e.g.*, Watson and Harrison, 2005), and broadly indicate crystallisation from cool, water saturated intrusive igneous systems (Fig. 1). Age *versus* X_{Al}^{Zrc} (Fig. 2) shows an increase in both the number of high Al (>4 ppm Al) and the average Al content of zircons beginning by ~3.6 Ga (Fig. 2).

Discussion

Al-in-zircon as a proxy for peraluminosity. X_{Al}^{Zrc} is influenced by melt composition, temperature and water content ($Al^{3+} + H^+ =$ Si⁴⁺) (Trail *et al.*, 2017; Wang and Trail, 2019). These parameters will co-vary in a magmatic system, and a magmatic liquid cooling along a liquid line of descent will generally increase peraluminosity and water content with decreasing temperature (e.g., Blatter et al., 2013). However, JH zircons record a narrow range of temperatures (Fig. 1). These temperatures indicate crystallisation from a cool, water-rich silicic magma which suggests that variations in water activity play a subordinate role in regulating X^{Zrc}_{Al}. The weak correlation between Al and Ti (Fig. S-5) suggests T also plays a subordinate role to composition in regulating X_{A1}^{Zrc} within this population (Trail et al., 2017). The dominant role of composition is further demonstrated in granitic zircons from the Lachlan Fold Belt (LFB), the type locality of I- and S-type granitic rocks [defined here as granitic rocks known to primarily form through igneous processes (I-type) versus those formed to a measurable extent by melting of metasedimentary precursors (S-types); e.g., Chappell, 1999]. It should be noted that I-type magmas are often weakly peraluminous (ASI < 1.1),

and as such bulk peraluminosity is not used here to define I- versus S-type magmas. In the LFB, even though Ti content of I-type zircons is slightly higher on average than S-types, the latter have significantly higher X_{AI}^{Zrc} (Fig. S-4)

Increasing peraluminosity results in increased alumina activity and a subsequent increase in X_{Al}^{Zrc} . For granitic zircons of the LFB, X_{Al}^{Zrc} exceeds 4 ppm when the bulk composition becomes peraluminous, likely reflecting this increase in alumina activity in the melt (Trail *et al.*, 2017). We use this 4 ppm value to discriminate between zircons that likely crystallised from peraluminous melts. It is worth noting that ~80 % of JH zircons have Al contents lower than the LFB I-types (Fig. 2b), which reflects the lower crystallisation T observed in the Ti content (Fig. 1c). Thus, this LFB based peraluminous JH zircons.

In the JH, there is a subtle but significant increase in X_{Al}^{Zrc} with decreasing age (Fig. 2b). The >3.8 Ga population yields X_{Al}^{Zrc} similar to I-type granitoids. In the 3.8–3 Ga population, likely peraluminous zircons (>4 ppm Al) have an average $X_{Al}^{Zrc} = 8.5$ ppm compared to $X_{Al}^{Zrc} = 5.5$ ppm in the >3.8 Ga group and $X_{Al}^{Zrc} = 7.8$ ppm in I-types. As the JH is a metasedimentary rock, this increase is akin to sedimentary mixing of metaluminous [ASI < 1, molar Al₂O₃/(Na₂O + K₂O) > 1] and peraluminous protoliths seen in modern sedimentary systems (Fig. S-7).

Origin of peraluminous zircons. The limited number of mechanisms to generate peraluminous melts makes their appearance in the JH record a useful event for interpreting





Figure 2 (a) Age *versus* Al concentration for JH zircons, error bars are 2σ . Zircons with X_{AI}^{2rc} below detection limit (b.d.l.) are indicated at base of the diagram. Horizontal dashed line at 4 ppm Al defines zircons that likely formed from peraluminous melts. (b) Cumulative probability diagram of I- and S-type LFB (Trail *et al.*, 2017) and age binned JH zircons. Numbers in inset are average X_{AI}^{2rc} for $X_{AI}^{2rc} >$ 4 ppm zircon populations from each group.

tectonic shifts on the early Earth. Experiments and investigations of modern magmatic systems have demonstrated multiple mechanisms to produce peraluminous magmas, including: (1) late stage differentiation of silicic "I-type" granitic magmas, (2) melting and incorporation of metasedimentary material, and (3) partial melting or fractionation of hydrous mafic parent material at >7 kbar.

Late stage differentiation of metaluminous I-type bulk compositions can produce weakly peraluminous melts. However, this peraluminosity yields a negligible amount of zircon (Trail *et al.*, 2017). Similarly, I-type systems can also evolve toward weakly peraluminous bulk compositions (Chappell, 1999 and sources therein), but peraluminous I-type zircons have similar compositions to their metaluminous counterparts (Fig. 3).

On Earth today, the most volumetrically significant way to produce strongly peraluminous melts is through melting of weathered sediments (*e.g.*, psammites, metapelites) (Patiño Douce and Johnston, 1991). Based on a P *versus* REE (or Dy) discrimination diagram that records the increase in P content of apatite saturated peraluminous melts, zircons from the Jack Hills have been dismissed as being derived from peraluminous melts (Burnham and Berry, 2017). However, there is significant overlap at low P content between peraluminous S-type and metaluminous and weakly peraluminous I-type granitoids (Fig. 3). Although the JH zircons do not exhibit high P-Dy signals, the overlap at low P content between S-type and I-type zircons makes it difficult to rule out metasediments as a source of some JH zircons, and does not discount previous work suggesting a potential sedimentary source for some JH zircons (Bell *et al.*, 2015).

In lieu of metasediment melting, moderate pressure fractionation of hydrous mafic material could produce peraluminous zircons in excess of those found in modern I-type granitoids. At pressures greater than ~7 kbar, hydrous mafic magmas can produce alumina-rich melts *via* crystallisation of Ca-rich pyroxene and suppression of plagioclase crystallisation (Müntener and Ulmer, 2006; Blatter *et al.*, 2013), and at pressures within the garnet stability field (≥ 10 kbar), incongruent melting of amphibole and biotite in metaluminous bulk compositions can produce alumina-rich melts and pyroxene (Chappell *et al.*, 2012). These moderate pressure processes lead to the generation of peraluminous melts at lower bulk SiO₂ (~60 wt. % SiO₂ *versus* ~70 wt. % SiO₂) than modern arcs magmas (Fig. 4), which in turn could yield a higher proportion of peraluminous zircons than modern arcs.

Implications for early Earth geodynamics. The distribution of X_{Al}^{Zrc} of JH zircons >3.8 Ga is similar to modern I-type magmas (Fig. 2), whose intermediate compositions overlap in SiO₂-ASI space with >4 Ga granitoids from the Acasta Gneiss Complex (AGC) that have been interpreted as crystallising from an environment akin to modern oceanic plateaus (Reimink *et al.*, 2014) at 3–4 kbar (Fig. 4). On the ancient Earth, this is consistent with crystallisation in a vertical tectonic regime prior to 3.8 Ga, and is further supported by other chemical similarities between pre-4 Ga AGC and JH zircons (Reimink *et al.*, 2019a).

Pre- and post-3.8 Ga JH and I-type populations cross the 4 ppm Al threshold at roughly the same cumulative probability (~90 %). However, the average Al content of the likely peraluminous post-3.8 Ga zircons is greater than either the pre-3.8 or I-type populations. This potentially indicates a different and strongly peraluminous source to the 3.8-3 Ga group. Regardless of whether the peraluminous zircons are derived from metasedimentary sources or moderate high pressure fractionation processes, both scenarios likely require a geodynamic shift toward horizontal tectonics. For sediment melting, weathered continental material is required to be brought to depths great enough to be melted. For deep mafic melting, hydrated near surface mafic material needs to be brought to moderate pressures (≥7 kbar) in order to melt and fractionate. Although it is possible for hydrated mafic material to be brought to depths through vertical tectonic processes, the relative short timescales invoked to account for the shift in ϵ^{176} Hf observed from JH and other ancient zircons (Bauer et al., 2020), similarities in trace element content between JH and AGC zircons (Reimink et al., 2019a), and the onset of peraluminosity indicate a shift toward a horizontal tectonic regime.

Whereas peraluminous granitic rocks dominate the bedrock geology in the AGC at ~3.6 Ga (Reimink *et al.*, 2019b), the peraluminous zircons that emerge in the JH record at ~3.6 Ga are only a fraction of the total zircon population (~10 %). This could indicate preservation bias of the peraluminous rocks of the AGC, preservation of more diverse protoliths in the detrital JH zircons, or a globally heterogeneous distribution of crust-forming processes *ca*. 3–3.6 Ga. Regardless, the emergence of significant peraluminous zircons in the JH conforms with observations from other ancient crustal systems (*e.g.*,



Figure 3 (a) Histograms of P/Dy ratios and (b) scatterplot of P and Dy concentrations in zircons from I-type and S-type LFB granitoids compared with likely peraluminous ($X_{AI}^{Zrc} > 4$ ppm) and non-peraluminous JH zircons. On the whole, peraluminous zircons from the JH have higher P/Dy ratios than non-peraluminous JH, and their median value of P/Dy is higher than that of peraluminous I-types. LFB data from (Burnham and Berry, 2017).



Figure 4 SiO₂ versus ASI for common magmatic rocks and melts. LFB and Tuolumne Intrusive Suite (TIS) intrusive rocks binned in 1 wt. % SiO₂ increments, error bars are 1 σ . I-type granitic rocks from the LFB (Griffin *et al.*, 1978; Chappell and White, 1992) and intrusive rocks from the TIS (Bateman *et al.*, 1984; Ratajeski *et al.*, 2001) become peraluminous at ~70 wt. % SiO₂, whereas hydrous basalts fractionated at depths \geq 7 kbar can produce melts that become peraluminous at ~60 wt. % SiO₂, unlike those fractionated at lower pressures (Blatter *et al.*, 2013).

AGC, Pilbara Craton; Bauer *et al.*, 2020) of a geodynamic shift toward horizontal tectonics by at least ~3.6 Ga.

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Additional Information

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Supplementary Information

The Supplementary Information includes:

- Extended Methods
- Supplementary Tables S-1 and S-2
- Supplementary Figures S-1 through S-8
- Supplementary Information References

Extended Methods

Whole rock samples were crushed to a sand using a BICO chipmunk jaw crusher with steel plates followed by a BICO pulveriser fitted with alumina disks. The resulting sand was screened using a water wash step to remove clays and fine particles, passed with a hand magnet to remove magnetic particles, and was run through a Frantz magnetic separator. Finally, methylene iodide was used to procure a concentrated zircon separate.

Zircons separates were mounted in epoxy and polished through their cores using successive 3 and 1 µm SiC lapping film, followed by 0.1 µm Type A diamond lapping film and a final polish using colloidal silica to flatten the surface of the zircons. Several epoxy mounts were also treated with an additional weak hydrofluoric acid surface treatment to facilitate dissolution of inclusions and metamict regions within zircons. While not necessary for quality analyses, this step did generally produce a higher proportion of clean laser ablation analyses than non-treated samples. Prior to analyses, each zircon was inspected in optical and transmitted light to ensure that analytical spots were not placed on metamict regions of zircons or on inclusions.

Sectioned zircons were analysed on an Agilent 7900 quadrupole inductively coupled plasma mass spectrometer (ICPMS) at the University of Rochester, fitted with a Cetac (Photon Machines) Analyte G2 193 nm laser ablation (LA) unit with a HelEx 2 volume chamber sample cell. Trace element data was collected with a 20 µm circular spot for analytical durations of 20 s with a 7 Hz pulse rate and a laser energy of 6.75 J/cm² (Wang and Trail, 2019). The number of masses analysed was minimised in order to increase the signal for the masses of primary interest. Masses analysed include: ⁷Li, ²³Na, ²⁴Mg, ²⁷Al, ³¹P, ⁴⁹Ti, ⁵⁷Fe, ¹⁴⁶Nd, ¹⁴⁷Sm, ¹⁶³Dy, ²⁰⁴Pb, ²⁰⁶Pb, and ²⁰⁷Pb (see Supplementary Table S-2).

Zircon U-Pb ages were determined following similar procedures to those described in Trail *et al.* (2017) Epoxy mounts were loaded into the HelEx 2 volume chamber and the He flow was set to 0.6 L/min in the chamber, and 0.2 L/min in the HelEx arm. The laser energy was set to 7 mJ (fluence = 11.8 J/cm^2), with a pulse rate of 10 Hz. A circular spot of 25 µm was selected for all analyses. For each analysis, background counts were collected first (~30 s), followed by 20 s of ablation, and then a 30 s washout period before moving on to the next zircon. Zircon ages were standardised against the AS-3 geochronology standard (Paces and Miller, 1993), and also monitored with a secondary in-house



standard, Kuehl Lake, believed to be from the same locality as the international zircon standard 91500 (Trail *et al.*, 2018). We analysed ²⁰²Hg, ²⁰⁴Pb, ²⁰⁶Pb, ²⁰⁷Pb, ²⁰⁸Pb, ²³²Th, and ²³⁸U. The integration times for ²⁰⁶Pb and ²⁰⁷Pb were set at 30 ms, and those for the other isotopes were set at 10 ms for each cycle. All data reduction, and correction for downhole Pb-U fractionation was done with the Iolite 3.32 software package (Paton *et al.*, 2018).

After chemical analyses, select zircons were also imaged using the panchromatic cathodoluminescence (CL) detector on the JEOL 8530F electron probe microanalyser (EPMA) at the Smithsonian Institution (Fig. S-1) in order to observe internal chemical zonation and to ensure that both age and chemistry laser spots targeted similar regions of the zircon.

Obtaining primary Al concentrations from the zircons was one of the main goals of this study. Al is known to be a contaminant in zircon analyses and is often used to monitor contamination in SIMS analysis (Piazolo *et al.*, 2016; Lyon *et al.*, 2019). Therefore, care was taken to ensure the filtering techniques described above eliminate non-primary Al measurements from consideration. There has been some discussion in the literature over the precise value of LREE index to use for filtering zircon compositions (Bell *et al.*, 2019). In our sample set, filtering of data for Fe and discordance prior to LREE-I filtering results in similar results, regardless of the LREE filter occurring at 30 or 50 (Fig. S-3).

Supplementary Tables

Table S-1Sampling locations for metasedimentary rocks of the Jack Hills presented in this study.

Sample	Easting	Northing
T136	499199	7106296
Т139	499207	7106283
T140	499193	7106279
T145	499235	7106170
T146	499231	7106157
T147	499245	7106139
T148	499253	7106125
T152	499274	7106030

Table S-2Sample numbers, ages, and chemical information for individual zircon crystals within the Jack Hillsmetamorphic belt.

The data table is available for download (Excel file) at https://www.geochemicalperspectivesletters.org/article2114.



Supplementary Figures



Figure S-1 Cathodoluminescence images of select Hadean and/or likely peraluminous zircons from the Jack Hills greenstone belt. 25 μ m spots were used for dating, and 20 μ m spots were used for chemical analyses. Reported errors on ages are 2 σ . Scale bars are 10 μ m.





Figure S-2 Representative time-resolved LA-ICPMS measurements for ²³Na, ⁵⁷Fe and ²⁷Al relative to ²⁹Si. Dashed boxes are the analytical window used to calculate trace element concentrations in the zircons. Care was taken to avoid regions of the time-resolved spectra that contained ephemeral spikes in elements that are likely caused by laser pits ablating impurities during analysis.



Figure S-3 Age versus X_{Al}^{Zrc} for JH zircons showing the change in population as a function of the filtering steps applied, including both the LREE-I 30 (Bell *et al.*, 2016) and LREE-I 50 filters (Bell *et al.*, 2019).





Figure S-4 Ti and Al concentrations in granitoid-hosted zircons from the Lachlan Fold Belt (LFB, Trail *et al.*, 2017), showing the dominant role of melt composition over temperature in regulating Al content in zircons in the LFB. As a proxy for temperature, Ti content in S-type zircons is similar to that of I-type zircons, indicating similar crystallization temperatures. However, S-type zircons crystallised from moderately to strongly peraluminous melts contain higher Al concentrations.





Figure S-5 Ti and Al concentrations in zircons from the Jack Hills, with little evidence of correlation between Ti (proxy for temperature) and Al content.





Figure S-6 Comparison of P-Dy and P-REE+Y plots, showing broad similarity between the two plots. Dy is used in thus study as a proxy for the a total REE plot (Burnham and Berry, 2017) as only a limited number of elements were analysed in the present study to maximise the signal from elements of interest during analysis.





Figure S-7 (Left) Al concentrations in quartz grains from sediments at the mouth of the Bega River, NSW, Australia compared with regional bedrock I- and S-type granitoids, showing mixed I- and S-type signals in the sediments that reflect the diversity of bedrock in the region (Ackerson *et al.*, 2015). (**Right**) JH zircons show a similar mixing between peraluminous and non-peraluminous sources, starting noticeably in the 3.5-3 Ga bin.





Figure S-8 Random subsampling of 3.5-3 Ga JH zircons. The large number of zircons from the 3.8-3 Ga bin (n = 332) compared to the 4.5-3.8 Ga (n = 66) makes it is possible that peraluminous signal in the Hadean-aged bin may have been missed by the limited number of Hadean-aged zircons measured. The 3.8-3 Ga bin was randomly subsampled to n = 66 zircon populations as a test of sample-number bias. At the 95 % confidence level using a Kolmogorov-Smirnov Test, only 1 of the 10 random subsamples was statistically similar to the 4.5-3.8 Ga bin, whereas all random subsamples were statistically similar to the 3.8-3 Ga bin. This suggests it is unlikely that under-sampling of the Hadean population failed to record a peraluminous signature.

Supplementary Information References

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