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Wriju Chowdhury (wchowdhu@ur.rochester.edu)
University of Rochester
Dustin Trail
University of Rochester https://orcid.org/0000-0003-3505-4351
Martha Miller
University of Rochester
Paul Savage
University of St. Andrews

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Eoarchean/Hadean melts reveal arc-like trace element and isotopic chemistry

Wriju Chowdhury[1*], Dustin Trail[1], Martha Miller[1], Paul Savage[2]

[1] Department of Earth & Environmental Sciences, University of Rochester, Rochester, NY 14627, USA.

[2] School of Earth and Environmental Sciences, University of St Andrews, Irvine Building, Scotland KY16 9AL, UK

*Corresponding author: Wriju Chowdhury (wchowdhu@ur.rochester.edu)

Summary (193 words)

Constraining the lithological diversity and geodynamic regime(s) of the earliest Earth is critical to understanding how our planet has evolved, and the discovery of Hadean- and Eoarchaean-age zircons has allowed us to directly analyse material from these eons. Here we use Jack Hills Zircon (JHZ) (3.3 - 4.2 Ga) analyses coupled with new experimental partitioning data to model the SiO_2 wt.%, Si+O isotopic composition, and trace element (TE) contents of their parent melts; i.e., the composition of the Earth's earliest crust. Comparing our derived JHZ parent melt Si+O isotopic compositions ($-1.96 \leq \delta^{30}Si_{NBS28} \leq 0.39$ ‰; $5.28 \leq \delta^{18}O_{VSMOW} \leq 10.03$ ‰) to younger continental crust lithologies, we conclude that the chemistry of the JHZ parent melts was influenced by the assimilation of serpentinites, cherts and silicified basalts, followed by igneous differentiation, leading to the formation of intermediate to felsic melts in the early Earth. Trace element measurements also show that the formational regime had an arc-like chemistry, consistent with the presence of mobile-lid tectonics in the Hadean. Moreover, based on the similarity of isotopic data derived for Eoarchaean and Hadean melts, we propose these continental-crust forming processes operated continuously, and uniformly, from the Hadean into the Archaean.

Main (2338 words)

The paucity of terrestrial geological samples from the Hadean and Eoarchaean means that making direct inferences about the geochemical and geodynamic nature of the first 500 million years of our planet is

fraught with controversy. Many researchers have made implicative arguments about the Hadean through the chemical and isotopic analysis of Hadean zircons and their mineral inclusions. This includes inferences about the types of geodynamic regimes and geological processes that could generate silicic melts (i.e. the earliest continental crust), hydrosphere-lithosphere interactions, and possible mobile-lid tectonics during a time when life may have evolved on our planet[1-7]. Even with access to ancient zircons, the presence or absence of mobile lid tectonics in the Hadean remains a vexed question, with arguments derived to support both end-members. While many have proposed a stagnant-lid for the Hadean Earth based on Hf, O and Nd isotopes [8-11] there are compelling arguments based on geophysical modeling, Nd isotopic evolution models, trace element chemistry of Hadean zircons and Ti isotopic values of the Acasta Gneiss rocks that point toward a mobile-lid regime on the early Earth [6, 12-17]. Ultimately, these end-member models based on contradictory, but empirical, observations need to be reconciled to provide us with an accurate view of early Earth's geological history.

Here we use in-situ analyses of Jack Hills Zircons (JHZs), to place quantitative constraints on the chemistry of their parental magmas, in terms of their trace (TE) and rare-earth (REE) elemental concentrations, SiO_2 content, and Si+O isotopic values. Quantifying SiO_2 content and Si+O isotopes are especially important given that they constitute ~75 wt. % of rock-forming elements in the crust and constitute a majority of the weathered and eroded material that makes up a significant portion of the oceans' nutrient budget. The degree of polymerisation of Si-O bonds also correlates with the relative density of the continental and oceanic crusts which controls long-term continental stability and rock-recycling. Additionally, we also explore the Al content of our zircons which is the third most abundant element in the crust and thus gives us insight into the aluminosity of the parent melts. Aluminosity of melts is of interest because it implies the incorporation of eroded stable subaerial crustal material (See Supplementary information).

<u>JHZ parental melt modelling</u>

To derive the SiO_2 content and Si+O isotopic values of our zircons' parental melts, we first model their parental melt trace element chemistry by combining in-situ zircon analyses and new experimentally-derived partition coefficients. Then, using melt Th/Y ratio as a proxy for SiO_2 content (whereby Th/Y was calibrated to melt wt% SiO_2 using whole-rock data of lithologies from the GEOROC database), we calculate the wt% SiO_2 concentration of JHZ parent melts[6]. The JHZ crystallization temperatures were estimated using the Ti-in-zircon thermometer [18]. The derived melt temperature and SiO_2 content both control zircon-melt Si and O isotope fractionation factors [19, 20]. The fractionation factors combined with in-situ zircon Si and O isotopic measurements are used to derive the melt Si and O compositions. Using this approach, we estimate the Si and O isotopic melt values for each individual Hadean and Eoarchaean zircon analysed, which we use as a petrological descriptor of early Earth crustal lithologies. Specifically, individual JHZ melt Si+O isotopic values can identify if supracrustal or altered sources, such as clays, shales, serpentinites, cherts, and/or silicified pillow basalts could have been involved in the formation of the aforementioned melts (while O isotopes in serpentinites have been well studied, Si isotopes for the same are not and so we also present new Si isotopic data from modern serpentinites). Ultimately, each JHZ analysis provides constraints on a parental melt's SiO_2 wt%, TE/REE, and Si/O isotope compositions, which allows us to infer the possible lithological and tectonic character of these zircon-bearing Hadean rocks.

JHZ parent melt silica content

Silicate melt Th/Y are well correlated with the SiO_2 content of whole rocks[6] (details and caveats discussed in Online methods), which we use to derive the melt silica content. As noted, we require the SiO_2 content of the Eoarchean/Hadean melts to derive the isotopic chemistry of the same. We measured the concentration of Th and Y of JHZs as a proxy for the melt Th and Y concentration. For these zircon data to be useful, we require zircon-melt partition coefficients (i.e., concentration in zircon divided by concentration in melt; $D_{element}^{zrc/melt}$) to constrain the TE/REE chemistry of the detrital JHZ melts. We conducted new zircon-melt partitioning experiments in an alkali- and water-bearing system and report D-

values for Th and Y of 0.72 ± 0.19 (2 s.e.) and 2.90 ± 0.61 respectively, which were used to derive the Eoarchaean and Hadean melt Th and Y concentrations (See online methods for details; Table ST1). Our calculated Th and Y concentrations were combined with an empirical calibration (See Online methods) to estimate melt SiO_2 (Fig 1; Table ST2). The calculated SiO_2 contents range from 51 to 76 wt% with an average value of 62 ± 11 wt% (2 s.d.) which suggests that most of the melts in question are between intermediate and felsic. In fact, all our accepted analyses except for two are above 55 wt% SiO_2 (15 of 84 rejected; see Online methods for rejection criteria). Finally, we used the Ti-in-zircon crystallization thermometer[18] to determine zircon crystallization temperature (Fig S2). The crystallization temperatures

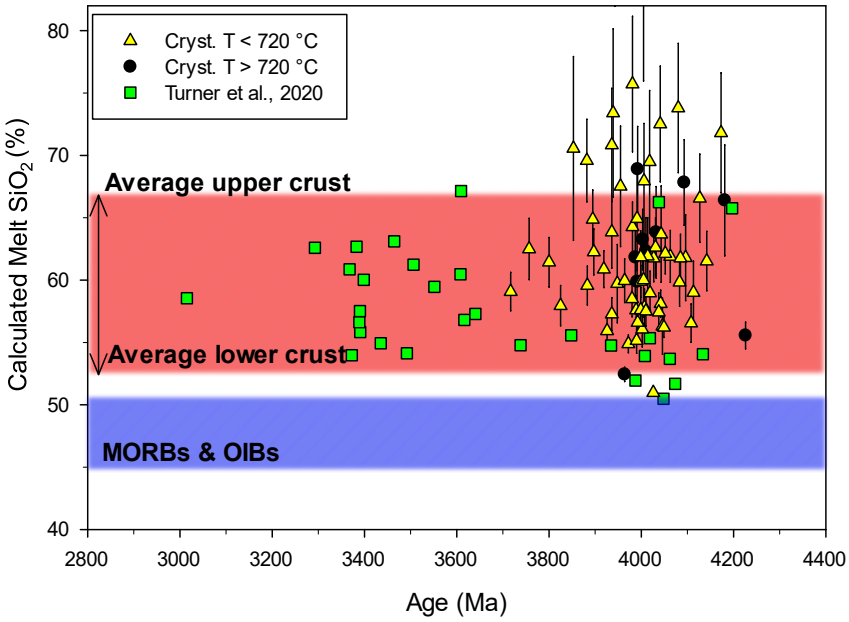

Figure 1. *Calculated melt SiO_2 wt% of Hadean and Eoarchaean melts shown against zircon ^{207}Pb-^{206}Pb age. The melt SiO_2 contents are calculated using new trace element partition coefficients reported here and are subtly higher (62 ± 11 wt% (2 s.d.)) than those reported in **[6]** (58 ± 12 wt% (2 s.d.)), who used D-values reported by **[33]**. **Field legend:** Red field: The upper bound is the average upper continental crust and the lower bound is the average lower continental crust. Blue field: All MORBs(Mid-Oceanic Ridge Basalts) and OIB (Ocean Island Basalt) values. The upper and lower crust values are from **[34]**. MORB and OIB values from **[29]**. Error bars are 2 s.e. (Details in Online Methods). Two zircons with apparent SiO_2 wt% melt values of 99.5% and 91% are not shown.*

range from 614°C – 774°C with an average of 685 ± 68 °C (2 s.d.) (Fig. S2(B)). Figure 1 and S2(B) both indicate that most of our JHZs crystallized from water-saturated felsic melts. With these parameters defined, we can derive the melt $\delta^{30}Si$ and $\delta^{18}O$ values.

JHZ parent melt isotopic content

We performed in-situ coupled $^{30}Si/^{28}Si$ and $^{18}O/^{16}O$ SIMS measurements on our JHZs (Fig S1; Table ST3). These measurements are necessary to derive the parent melt isotopic content (Table ST4) using fractionation factors. Along with temperature, the primary melt property that affects mineral-melt fractionation factors is the melt silica content and the resulting polymerization of Si-O bonds [19, 21]. Thus, the derived chemical parameters from the previous section (SiO_2 wt % and crystallization T) allows us to obtain zircon-melt fractionation factors for Si and O isotopes using experimental and empirical calibrations[19, 20] (see online methods), and thereafter the Si and O isotopic composition of the melt. These modelled melt data are far more implicative for deconvolving the petrogenesis of Hadean melts, as we can now compare these isotopic compositions to possible source lithologies (rather than just zircon data alone, see Extended data figures).

JHZ parent melt constituents and formational regime

In a $\delta^{30}Si$ vs. $\delta^{18}O$ plot, we compare our parent melt Si and O isotopic values to potential source lithologies, as well as end-member mantle reservoirs (EM1 (Enriched Mantle 1), EM2 (Enriched Mantle 2) and HIMU (High-μ)) and Archaean komatiites (Fig. 2) to quantify possible interacting reservoirs and lithologies that could have created our JHZ melts. An important assumption is that the Phanerozoic and more recent lithologies plotted in Fig. 2 are chemically analogous to similar rocks formed in the Archaean and Hadean. The similarity of Archaean komatiites to modern serpentinites from ODP holes 1268A and 1274A (Table ST4) as well as mantle reservoirs indicate hydrothermal processes that alter mantle lithologies alone cannot account for the JHZ melt values and thus lithologies with elevated $\delta^{30}Si$ and $\delta^{18}O$

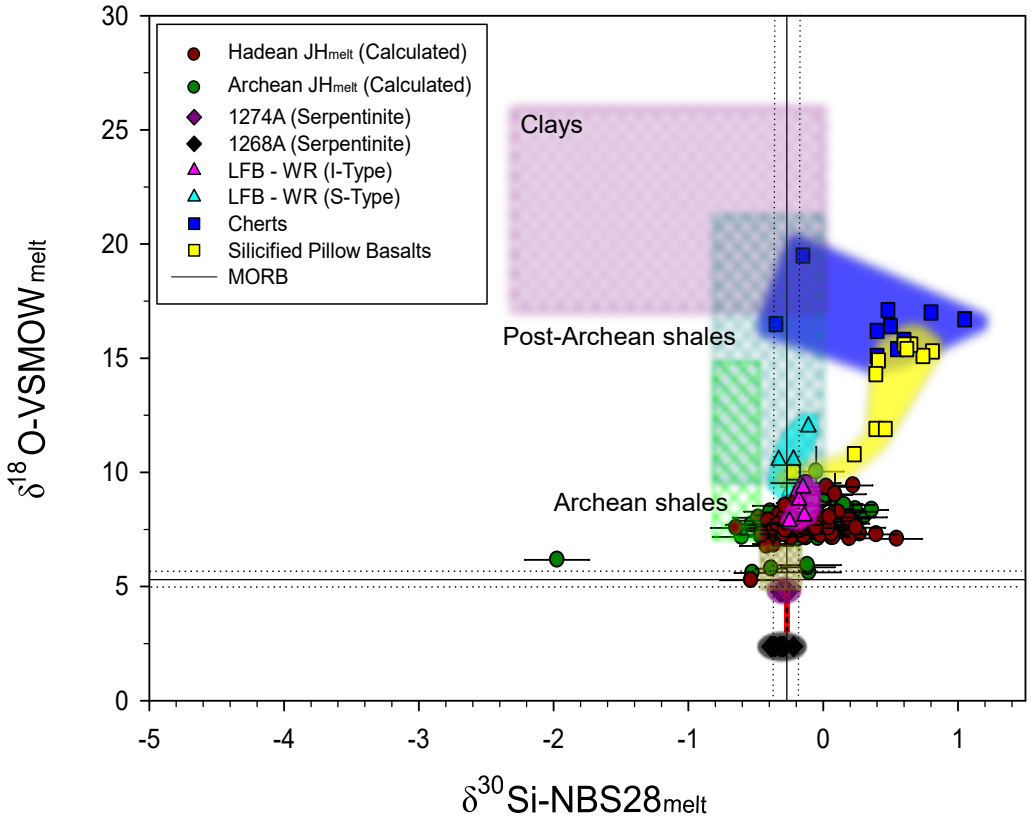

Figure 2. *$\delta^{30}Si$ and $\delta^{18}O$ (±2 s.e; see online methods.) values of derived Hadean and Archaean melts in comparison with Archaean cherts/ silicified pillow basalts from Barberton, South Africa [35] and modern ODP serpentinites ($\delta^{18}O$[36]). Also shown are clays (dark pink box; $\delta^{30}Si$[37]; $\delta^{18}O$[38]), shales (Dark Cyan and green boxes; $\delta^{30}Si$ [39]; $\delta^{18}O$[40]) OIBs (olive green box; $\delta^{30}Si$ of HIMU (Mangaia, Cook Islands), EM1(Pitcairn islands) and EM2 (Samoa))[41]; $\delta^{18}O$ of EM1 and EM2[42]; $\delta^{18}O$ of HIMU[42] (Olivine phenocrysts[43], $\Delta^{18}O_{melt\text{-}olivine}$[44]) representing HIMU, EM1 and EM2 mantle reservoirs and Archaean komatiites (Red bar; $\delta^{30}Si$:[126], $\delta^{18}O$[45, 46]). JH_{melt} $\delta^{18}O$, serpentinite error bars are smaller than the symbols. The assimilation of these lithologies was also hinted at in just the JHZ isotopic data (Fig. S1). Data for MORB[47].*

need to be invoked. We do so by comparing our calculated parent melts to modern granitoids from the Lachlan Fold Belt (LFB)[21], clays, modern to Archaean shales, Archaean cherts, and Archaean silicified pillow basalts. The mafic lithologies, sediments and cherts/silicified pillow basalts seem to suggest that these might be possible rocks that could have re-melted to create the composite melts from which the JHZs crystallized. The overlap between our melts and the Phanerozoic granitoids implies that some early Earth melts share chemical similarities with modern I-type granitoids. Although, when considering the isotopic values (low $\delta^{18}O$) for Archean shales, sediments like this could very well have been involved in the melting process, so S-type granitoid melts are also permissible when considering the isotope data.

If we compare the $\delta^{30}Si$ values of JHZ melts to recently reported values of Archaean rocks (Fig. 3A), there is overlap between our JHZ melts and Archaean cherts, silicified pillow basalts, TTGs, GMSs (Granite-Monzonite-Syenite) and komatiites, but the isotopic value of these Archaean rocks cannot explain the larger range in $\delta^{30}Si$ of all JHZ melts. They require lithologies with a large $\delta^{30}Si$ range[22 - 24] that may be assimilated to generate the melt values we derived. Archaean chert (Fig. 3A; grey crossed squares) is a possible lithology that might have been involved in shifting the $\delta^{30}Si$ values to greater-than mantle values given that, on balance, JHZ parent melt $\delta^{30}Si$ values are slightly heavier (Hadean: -0.13 ± 0.06 ‰; Archaean: -0.14 ± 0.09‰ (2 s.e.)) than the MORB reference ($\delta^{30}Si$ = -0.27 ‰). To test this observation, we performed two separate t-tests which compared the Hadean and Archaean $\delta^{30}Si$ values to the MORB reference and both tests showed that the Hadean and Archaean average $\delta^{30}Si$ values are statistically different from the MORB reference (see Online methods). This gives us a rare insight into

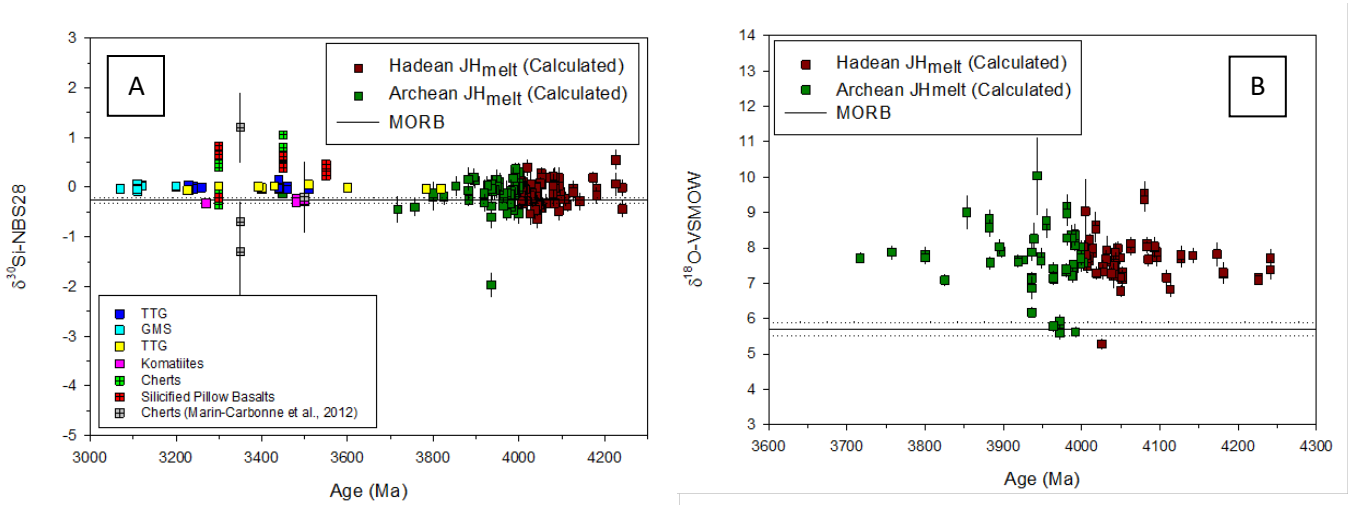

Figure 3A. *$\delta^{30}Si$ of JH parent melts vs. age. The parent melts have been grouped by age. JH melts are compared to Archaean rocks.* ***B.*** *$\delta^{18}O$ of JH parent melts vs. age. In both graphs, the calculated isotopic composition of the Hadean and Archaean melts are statistically identical.* ***Data sources****: Cherts/Silicified pillow basalts[23,36] (Green and red crossed squares; same data as Fig. 2 and Grey crossed squares); TTGs[25, 26]; GMS[25]; Komatiite[26]; MORB[47].*

what Hadean lithologies might have been. Similar arguments have been made to justify the $\delta^{30}Si$ values of Archean TTGs[25, 26].

Also, z-tests between the Hadean and the Archaean zircons (details in supplementary), show no statistical difference between the Hadean and Archaean melt values in either isotopic (Fig. 3A & B) system. This lack of a significant difference could imply that the geological processes and sources that gave the Archaean JH parental melts their Si and O isotopic values (and SiO_2 content) began operating during the Hadean.

Based on the inferences from our Si and O isotope models, sources involved in generating zircon-bearing melts probably include cherts, serpentinites and chert/silicified pillow basalts. The involvement of these source lithologies also imply the precipitation of cherts, low/high-T water-rock interaction to form serpentinites and silicification of pillow basalts, their re-melting, and the evolution of the composite melt to the point of zircon saturation all before the oldest zircon in our study crystallized (~4.2 Ga). A suitable tectonic regime thus needs to account for all these processes as well as the possible constituent lithologies. One possibility, among others[17], is a plate-boundary regime that some liken to a modern subduction zone[6] where our proposed lithologies are recycled and interact with mantle reservoirs. Some subduction zone properties implied by JHZs are that the JH parent melts may have been felsic, water-saturated, similar in its volatile content to modern island arcs and formed in a low-T (650-800 °C), high-P (>4 kbar) regime [2, 7, 17, 27]. As a test for this possible formational regime, we compare our JHZ melt TE/REE values to modern lavas.

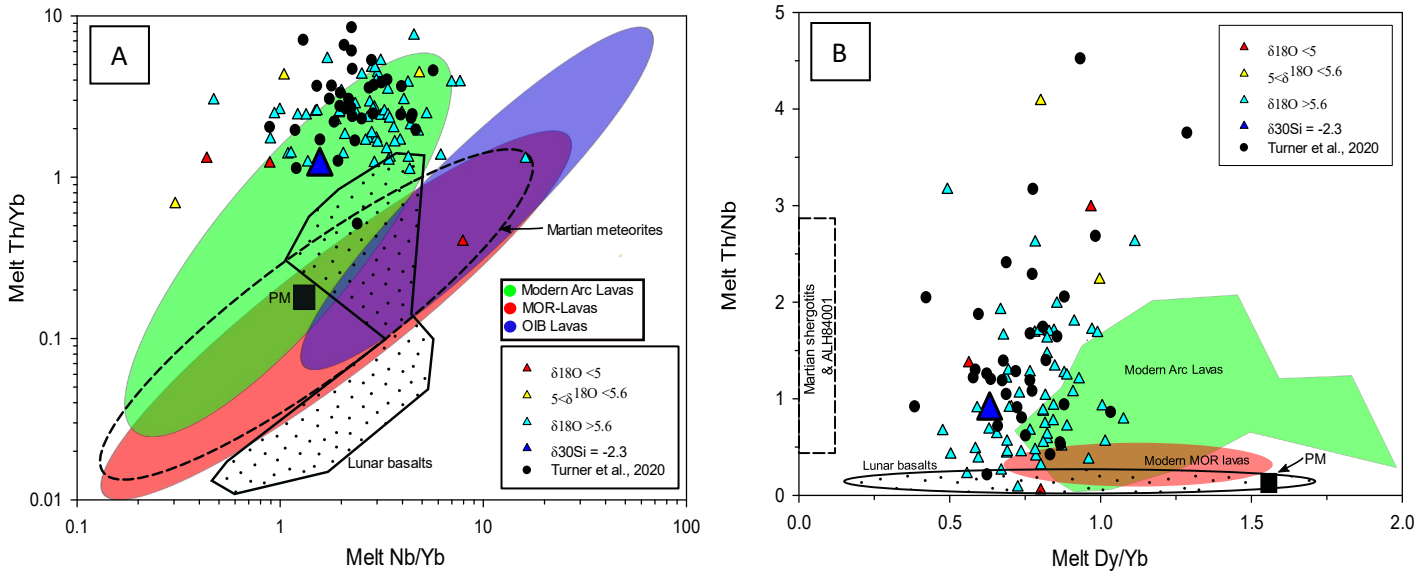

***Figure 4 A & B**: Comparison of elemental ratios of derived melt values between our JHZs (red, yellow, cyan and blue triangles) and those reported in **[6]** (black circles). **A:** Th-Nb proxy used to classify lavas from different regimes. Lunar and martian samples are shown as possible stagnant-lid regime analogues[48]. **Field legend: Ocean Island Lavas (Blue), MOR lavas (Red), Modern arc lavas (Green). Data sources: Terrestrial lavas [31]; Lunar basalts and martian meteorites[48] B:** Dy/Yb proxy for calc-alkaline differentiation as well as for restite garnet along with the Th/Nb proxy. **Data sources: MOR lavas [31]; Modern arc lavas [32] Martian meteorites [49, 50] Lunar basalts [51]. Primitive Mantle** (PM; Black square) [52]. Trace element melt contents were derived from zircon using newly derived partition coefficients (see Extended data figures). Errors for our data are given in Table ST2.*

With the melt TE/REE values derived using our new partition coefficients, we compared JHZ melts to melt values proposed by **[6]** and compared JHZs of this study and from **[6]** to lavas from modern regimes where crust is generated on Earth, on discrimination diagrams (Fig. 4 A&B). We have also shown lunar basalts and martian meteorites fields as possible analogues for stagnant-lid tectonic scenarios with a caveat that these extra-terrestrial regimes might be anhydrous. These diagrams have been used to fingerprint tectonic settings of rocks and minerals[28-32]. The Th/Nb proxy (Fig. 4A) is useful in differentiating between oceanic basalts and lavas with a continental input. Thorium and Nb have similar compatibility during partial melting of the mantle to form basalts, but continental input increases the Th content of melts. The Dy/Yb ratio (Fig. 4B) is used as a proxy for calc-alkaline differentiation as well as a proxy for garnet. SiO_2 % and Dy/Yb are negatively correlated while the crystallization of garnet increases the Dy/Yb ratio in the restite melt. Most of our melt values lie in the arc lava field in Fig. 4A, while in 4B

only some of them do so. They, however, do not fall in the MOR lava, lunar basalt or martian meteorite fields. This implies that our JHZs and those of **[6]** were created in a regime that has chemical similarities with modern arcs. We would like to mention that a system as complex as a subduction regime that comprises several reservoirs such as fluxing fluids, mantle wedges, subducting slabs, sediments, etc. may require multiple avenues of inquiry when attempting to define its signature chemistry and our study of TEs, combined with isotopic measurements, presents one such avenue. One of the melts with lower-than-mantle $\delta^{18}O$ zircon value falls in the mid-oceanic ridge field and so, may have originated in a setting analogous to this. However, there are a couple of melts with greater-than-mantle $\delta^{18}O$ that also fall in this field and some mantle-like $\delta^{18}O$ melts fall in the arc lava field. The majority of our datapoints are most definitely chemically distinct from lunar basalts or martian meteorites. Even so, we find that JHZs discussed here and in **[6]** strongly suggest formation in a regime with subduction zone-like chemical characteristics. This observation and the dissimilarities with lunar and martian rocks places specific chemical constraints on the JHZ formational environment and provides a limiting scenario about the geodynamics of the early Earth.

We propose that water-rock interactions took place at the Earth's surface to generate Hadean cherts, silicified pillow basalts and serpentinites. These lithologies were subsequently re-melted to generate felsic, peraluminous/metaluminous melts based on Th/Y ratio and Al content. Moreover, these interactions and re-melting processes gave the Hadean and the Archaean JHZs the same Si and O isotopic content and thus might have been uniformly operating across these two eons. Finally, whatever the precise details are of the regime that did create these zircons, our calculated WR compositions show clear chemical similarities between it and modern plate boundaries. We are proposing naturally and experimentally derived models that directly quantify the chemical and physical state of our planet during its evolution during the first 500 to 700 Myr when life might have emerged[3]. This period of our planet is important now; since we are exploring the habitability of other terrestrial-like planets some of which may be like the Hadean Earth.

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