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$^{238}U/^{235}U$ of zircons record the tectonic regime transition in crustal formation at 3.0 Ga

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Dear Editor,

The composition and formation mechanisms of Earth's continental crust represent fundamental puzzles in planetary evolution. Unlike other terrestrial planets, Earth's felsic-dominated crust is primarily generated at convergent margins via subduction-driven mantle melting. However, the timing of the onset of plate tectonics as the dominant mechanism for continental crust formation, and whether this mechanism operated throughout Earth's history, has remained vigorously debated.[1,2]

Archean continental crust mainly comprises tonalite-trondhjemite-granodiorite (TTG) suites, widely interpreted as products of hydrated basalt melting. Yet, their geodynamic origins are disputed. Two competing models have emerged: (1) a mobile-lid regime, invoking subduction-related processes such as slab melting and arc-like magmatism versus (2) a stagnant-lid regime, positing TTGs formation through the melting of overthickened mafic crust in tectonically stagnant settings.[1,3] Resolving this dichotomy is pivotal to constraining continental crustal origins and Earth's early tectonic transitions.

Zircon ($ZrSiO_4$), an excellent refractory mineral, can preserve robust records of original magmatic compositions even under high-grade metamorphism or partial melting, making it a critical archive for probing early crustal evolution. Uranium, a radioactive trace element enriched in zircon, not only provides high-precision geochronology via U-Pb systematics but also serves as a redox-sensitive tracer.[4] The U isotope composition ($\delta^{238}U$, defined as $[(^{238}U/^{235}U)_{Sample}/(^{238}U/^{235}U)_{CRM\text{-}145} - 1] \times 1000$ (‰), with CRM-145 as the international reference standard[4]) of zircon exhibits marked potential for tracking magmatic differentiation and redox fluctuations, given that the incompatibility of U and its sensitivity to oxygen fugacity (fO_2).[4] Thus, systematic investigation of secular $\delta^{238}U$ variations in zircons may unravel long-term magmatic trends, providing critical insights into the mechanisms and tectonic drivers of continental crust formation.

In this study, we compile U isotope data of magmatic zircons across multiple cratons, including Pilbara, Superior, North Atlantic, and East Antarctica, as well as detrital zircons from Jack Hills, spanning back to 4.2 Ga.[4] The zircon $\delta^{238}U$ distribution, displayed through box-and-whisker plots with 0.5 Ga binning intervals (Figure 1A), identifies 3.0 Ga as a transitional boundary. A Student's *t*-test confirms a statistically robust distinction between zircon populations before and after this boundary ($p < 0.0001$). Specifically, mean $\delta^{238}U$ increases markedly from -0.23 ± 0.05‰ (2SE; n = 30) in pre-3.0 Ga zircons to -0.10 ± 0.03‰ (2SE; n = 46) in post-3.0 Ga samples (Figure 1B). This U isotope composition shift likely reflects fundamental changes in felsic magmatism during the mid-Archean, offering novel insights into the mechanisms underlying early continental crust formation.

POTENTIAL MECHANISMS FOR $\delta^{238}U$ VARIATION

Weathering-driven preferential leaching of ^{235}U could drive ^{238}U enrichment in altered zircons,[4] but our dataset reveals an inverse trend. That is, pre-3.0 Ga zircons exhibit lower $\delta^{238}U$ compared with post-3.0 Ga zircons. This correlation with age suggests that the temporal evolution of U isotope signatures in zircon is not primarily governed by weathering, but rather reflects the physicochemical conditions during zircon crystallization. The elevated $\delta^{238}U$ signatures in younger zircons may be explained by two alternative mechanisms: (1) a shift in the U isotope composition of their magmatic sources, potentially involving incorporation of ^{238}U-enriched materials; or (2) an increase in the U isotope fractionation factor between zircon and melt during crystallization.

As a redox-sensitive element, U isotope behavior in surface environments is primarily governed by atmospheric-oceanic redox conditions. During the Archean, Earth's surficial systems remained largely reducing, lacking efficient reservoirs to enrich ^{238}U. This is attributed to the persistently reducing atmosphere-ocean system prior to the Neoproterozoic Oxidation Event (NOE, ~0.6 Ga), which significantly limited the scale of U geochemical cycling.[5] Although the Great Oxidation Event (GOE, ~2.4 Ga) raised atmospheric O_2 and enabled U^{6+} delivery to oceans, but U was quantitatively removed under reducing conditions, limiting isotope fractionation.[5] Sedimentary records show that prior to the NOE, the $\delta^{238}U$ of carbonate rocks and shales were similar to that of the continental crust.[4] Uniform $\delta^{238}U$ (≈ -0.3‰) across major geochemical reservoirs, including the depleted upper mantle, deep mantle reservoir, and altered oceanic crust, further supports the limited isotope heterogeneity in magma sources before the NOE.[4,5] These observations suggest that the anomalous increase in $\delta^{238}U$ in zircons after ~3.0 Ga is more likely attributed to a shift in the zircon-melt isotope fractionation factor, rather than the incorporation of ^{238}U-enriched material. In this study, we assume the melt from which zircons crystallized possessed a U isotope composition consistent with that of the continental crust (CC: -0.29 ± 0.03‰).[4] Accordingly, we infer that around 3.0 Ga, the zircon-melt U isotope fractionation ($\Delta^{238}U_{zircon\text{-}melt}$) increased from 0.06‰ to 0.19‰.

Uranium isotope fractionation between zircon and melt is closely linked to the magmatic physicochemical conditions, particularly crystallization temperature and fO_2. Global zircon datasets reveal a pronounced increase in the Ti-in-zircon temperatures, rising from ~620°C in the Hadean to ~900°C by the end of the Archean.[1] Since U isotope fractionation is governed by both nuclear volume and mass-dependent effects, it scales with 1/T and $1/T^2$, respectively.[6] This implies greater fractionation at lower crystallization temperatures. However, contrary to this expectation, zircons formed at higher temperatures display greater $\delta^{238}U$ offsets, suggesting that temperature alone cannot account for the shift in U isotope compositions around 3.0 Ga, and is therefore unlikely to be the dominant controlling factor.

REDOX CONTROL ON U ISOTOPE FRACTIONATION

Magmatic fO_2 may be a key factor controlling U isotope compositions in zircon. The speciation of U in silicate melts (U^{4+}, U^{5+}, and U^{6+}) is strongly dependent on redox conditions.[7] With increasing fO_2, higher-valence species (U^{5+} and U^{6+}) become more abundant, while U^{4+} diminishes.[7] This speciation shift directly influences U partitioning behavior: U^{4+} is more compatible with the zircon lattice, resulting in higher zircon-melt partition coefficients under reducing conditions.[7] Uranium isotope fractionation is primarily driven by the nuclear volume effect, whereby U^{4+} preferentially incorporate ^{238}U, while higher-valence species favor ^{235}U.[6] As a result, under high-fO_2 conditions, ^{238}U-enriched U^{4+} is preferentially partitioned into zircon, leaving the residual melt relatively enriched in ^{235}U. This redox-mediated fractionation mechanism provides a framework for interpreting $\delta^{238}U$ variations in zircon throughout geological history.

Based on the distribution of U oxidation states in silicate melts, U^{4+}/U^{6+} partition coefficients, and U^{4+}-U^{6+} isotope fractionation factors, the equilibrium U isotope fractionation ($\Delta^{238}U_{zircon-melt}$) can be estimated across varying fO_2 conditions (See https://doi.org/10.17632/ym4yxwtpjk.1 for details). The results reveal that $\Delta^{238}U_{zircon-melt}$ increases systematically with rising fO_2 (Figure 1C), suggesting that U isotope compositions of zircons may serve as a proxy for magmatic redox conditions. Accordingly, the global post-3.0 Ga shift toward higher $\delta^{238}U$ in zircon, with $\Delta^{238}U_{zircon-melt}$ increasing from ~0.06‰

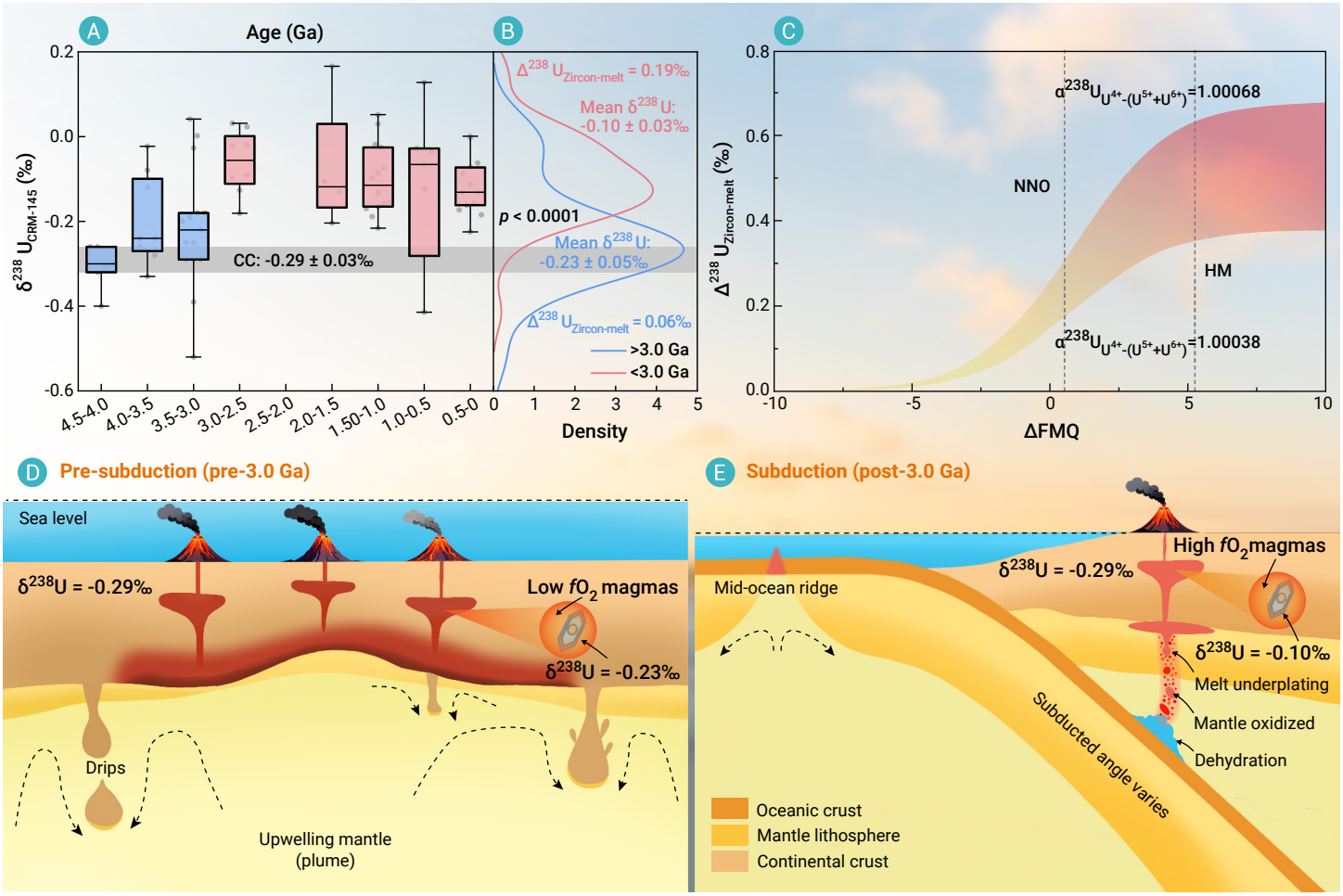

Figure 1. Zircon U isotope variations at ~3.0 Ga were governed by magmatic oxygen fugacity, reflecting shifts in crust-forming tectonic regimes (A) Box-and-whisker plots illustrating the distribution of zircon $\delta^{238}U$ grouped at 0.5 Ga intervals. Boxes represent the 25th-75th percentiles, with horizontal lines denoting median values. Whiskers extend to the full data range (0th-100th percentiles). (B) Kernel density estimates comparing $\delta^{238}U$ distributions before and after 3.0 Ga. (C) Calculated equilibrium U isotope fractionation between zircon and melt ($\Delta^{238}U_{zircon-melt}$), integrating U speciation, partitioning and valence-dependent isotope fractionation factors ($\alpha^{238}U_{U^{4+}-(U^{5+}+U^{6+})}$ = 1.00038~1.00068).[6,7] (D) Stagnant-lid regime: during incipient lithospheric cooling, mantle-derived melts ascend via plume. In rapidly cooled lithospheric regions, gravitational instability led to the foundering of lithospheric material as "drips". During this dynamic process, hydrated lower crustal rocks at varying depths underwent partial melting, generating TTG magmas. (E) Mobile-lid initiation: lithospheric breakup marks the onset of proto-subduction. Upwelling asthenosphere at mid-ocean ridge systems forms new oceanic crust, while convergent boundaries develop slab wedging, a hallmark of modern plate tectonics. Slab dehydration triggers metasomatic oxidation of the overlying mantle wedge, producing oxidized TTGs and subsequent K-rich magmas.

to ~0.19‰ (Figure 1B), is most likely a response to the elevated magmatic fO_2.

TECTONIC MODULATION OF MAGMATIC OXIDATION

Tectonic processes have played a fundamental role in modulating the oxidation state of magmas throughout Earth's history.[7] The evolution of fO_2 in felsic magmas reflects redox changes in magmatic systems across diverse tectonic settings. Modern observations show that arc magmas exhibit significantly higher fO_2 (ΔFMQ +1.4 ± 1.1) compared to mid-ocean ridge and intraplate magmas (ΔFMQ ~0 ± 0.5).[7] This contrast is primarily attributed to the influx of oxidizing agents, such as sulfate species and Fe^{3+}, from altered oceanic crust and overlying sediments into the mantle wedge above subduction zones. The widespread generation of oxidizing, water-rich calc-alkaline magmas during the Phanerozoic is correlated with the extensive development of an oxidized deep-ocean environment.[8] In contrast, the predominantly anoxic surface conditions of the Archean suggest that alternative oxidation mechanisms operated during this time.[8] Archean arc magmas may have been oxidized through processes involving water dissociation and hydrogen incorporation into orthopyroxene.[8] The hydrated ancient oceanic crust, with strong capacity in holding water during hydration, likely underwent extensive dehydration melting upon subduction into hotter Archean mantle.[1] This process may have provided a considerable flux of fluids, thereby playing a critical role in promoting the oxidation state of arc magmas. Such slab-derived deep-water cycle oxidation, operating independently of surface redox conditions, may have governed the formation of oxidized arc magmas in the Archean. By contrast, primitive magmas produced in other tectonic settings, including hotspots and mid-ocean ridge, produce hot, dry tholeiitic magmas with lower fO_2.[7] Although gravitational dripping has been proposed as a mechanism for early continental crust formation,[1] there is currently insufficient evidence to suggest that this process can significantly elevate magma oxidation states. Indeed, magmas generated under stagnant-lid conditions where gravitational dripping is prevalent, exhibit markedly more reduced characteristics compared to those from subduction-related tectonic environments.[8] Collectively, the consistent association between high water content and elevated fO_2 in arc magmas underscores a fundamental coupling among magma composition, oxidation state, and tectonic setting.

3.0 GA GEODYNAMIC TRANSITION: FROM A STAGNANT-LID TO A MOBILE-LID REGIME

Uranium isotope compositions of zircons reveal a marked increase in felsic magma fO_2 around 3.0 Ga, suggesting the onset of water-dominated magmatic evolution. This shift may reflect a fundamental geodynamic transition, from a stagnant-lid to a mobile-lid regime, characterized by the initiation of subduction and deep-water recycling (Figures 1D-E).[9] Supporting this interpretation is the widespread emergence of magnetite-series, potassic granitoids formed during syn- to late-tectonic stages across several Archean cratons.[1,8] Contemporaneous zircon Hf-O isotope records show a slowdown in continental growth rates,[1,2] while V isotope data from global diamictites

reflect a shift in upper crust composition from mafic to felsic dominance.[10] These changes align with peak upper mantle temperatures, suggesting that long-term mantle cooling may have driven this tectonic transition.[1] Notably, the fO_2 proxy $\delta^{238}U$ exhibits a narrow range over the subsequent 3.0 Ga (Figure 1A). This long-term redox stability suggests that subduction-driven hydrous magmatism, together with plate tectonics, has sustained a continuous geodynamic regime.

IMPLICATIONS FOR THE RISE OF ATMOSPHERIC OXYGEN IN EARLY EARTH

This study demonstrates that variations in U isotope compositions of zircons reveal a key shift in Earth's geodynamic evolution during the mid-Archean (~3.0 Ga), marked by the onset of plate tectonics. The establishment of subduction systems initiated large-scale deep-water cycling, which became a key driver in the generation of oxidized arc magmas. The involvement of water significantly enhanced magma differentiation, promoting the widespread production of felsic rocks.[3] More importantly, oxidized felsic magmas derived from subduction zones, characterized by low Fe and S contents and elevated $Fe^{3+}/\Sigma Fe$ ratios, consumed far less free O_2 during chemical weathering compared to mafic rocks. This differential weathering effect likely played a crucial role in facilitating the rise of atmospheric O_2 during the early Paleoproterozoic.[10] Additionally, subduction-induced mantle oxidation may have reduced the flux of reduced volcanic gases to the atmosphere. Together, these dual regulatory mechanisms may have jointly contributed to the triggering of the GOE.

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AUTHOR CONTRIBUTIONS

Jiaru Sheng: Investigation, Methodology, Visualization, Writing-original draft, Writing-review&editing. Yikang Quan: Writing-review&editing. Haochen Duan: Writing-review&editing. Dingsheng Jiang: Writing-review&editing. Wei Wei: Writing-review&editing. Fang Huang: Investigation, Conceptualization, Project administration, Writing-review&editing. All authors contributed to the manuscript and approved the final version.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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