

Secular evolution of rhyolites: Insights into the onset of plate tectonics

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Abstract. The emergence of plate tectonics is intimately linked to the stabilization of buoyant continental lithosphere, yet the timing of this transition remains contentious. Here, we analyze the secular geochemical evolution of rhyolites, a proxy for crustal differentiation, to constrain the onset of modern-style plate dynamics. A global compilation of 21,252 rhyolitic samples reveals statistically significant shifts in diagnostic geochemical indicators at ~2.7 Ga, most notably increased potassium (3–5 wt%) and intensified negative Eu anomalies ($\text{Eu}/\text{Eu}^* = 0.3\text{--}0.6$). These trends mirror Phanerozoic rhyolites and temporally coupled with supercraton assembly (Kenorland), peak crustal reworking rates, [seismic velocity structure indicative of felsic upper crust](#) and the oldest evidence of plate margin processes (e.g., passive margins, foreland basins). The ~2.7 Ga shift in rhyolite compositions, including elevated K_2O , pronounced Eu anomalies, and enriched Nd isotopes, reflects enhanced crustal reworking and the stabilization of rigid continental lithosphere. This transition is rooted in the long-term accumulation and maturation of tonalite-trondhjemite-granodiorite (TTG) suites, which are the primary building blocks of Archean felsic crust. [The estimated minimum upper crust seismic velocities for Archean cratons are systematically lower post-2.7 Ga, comparable to modern continental upper crust, indicating a more differentiated crust for Archean blocks since 2.7 Ga.](#) This period coincides with structural records of large-scale horizontal lithospheric motion and the establishment of interconnected plate boundaries, confirming the emergence of conditions necessary for sustained plate tectonics. The geochemical proxies in rhyolites provide quantifiable evidence for continental rigidization, complementing structural and isotopic archives of early plate dynamics. We propose that the ~2.7 Ga surge in evolved rhyolites marks the stabilization of rigid continental lithosphere—a prerequisite for sustained plate tectonics. This study reconciles conflicting models by linking crustal maturation to the earliest definitive records of convergent margins, establishing ~2.7 Ga as a pivotal transition in Earth's shift to modern geodynamics.

1 Introduction

Plate tectonics governs Earth's geochemical cycles, crustal growth, and biosphere evolution (Goldfarb et al., 2010; Liu et al., 2019; Russell et al., 2010; Wilson, 1968). However, the timing of its onset remains debated, with estimates ranging from the Hadean (>4.0 Ga) (Turner et al., 2020) to the Neoproterozoic (<1.0 Ga) (Stern, 2005). A critical prerequisite is the formation of thick, buoyant continental lithosphere capable of resisting mantle traction (Cawood et al., 2018; Stern, 2005), which provides mechanical resilience if subduction and collision occurred (Stern, 2005). Early Earth's hotter mantle favored mafic, denser

and ductile crust production (Herzberg et al., 2010; Korenaga, 2008; Tang et al., 2016), but by ~3.0 Ga, felsic components began to dominate, as evidenced by sedimentary proxies (Cai et al., 2023; Smit and Mezger, 2017; Tang et al., 2016; Tian et al., 2023). Although several lines of evidence suggesting the presence of rigid continental crust occurred as early as 4 billion years ago (Fig. 1), it still remains unclear to what extent the crustal maturation facilitated the operation of plate tectonics, especially considering the higher mantle temperatures during the Hadean and Archean eons (Herzberg et al., 2010; Korenaga, 2008).

Rhyolites, high-silica volcanic rocks ($\text{SiO}_2 > 68 \text{ wt\%}$), require melting of evolved crustal sources (Thompson and Connolly, 1995). They can form through two primary mechanisms: (1) partial melting of (meta-)igneous or (meta-)sedimentary crust, or (2) extraction of interstitial melt from long-lived crystalline mushes—mixtures of crystals and melt that represent the dominant reservoir for silicic magmatism (Ayalew et al., 2002; Bachmann and Bergantz, 2008; Bacon and Druitt, 1988; Cameron and Hanson, 1982; Grove et al., 1997). The latter process, involving melt segregation from mush zones at intermediate crystallinities (50–60 vol. % crystals), is particularly effective in producing voluminous, crystal-poor rhyolites (Bachmann & Bergantz, 2004; Hildreth, 2004). Importantly, the tectonic setting imparts distinct geochemical signatures: rhyolites from subduction zones (cold-wet-oxidized) typically exhibit U-shaped REE patterns due to amphibole and titanite fractionation, whereas those from mantle upwelling settings (hot-dry-reduced) show “seagull” REE patterns with deep Eu anomalies, reflecting plagioclase-dominated fractionation (Bachmann & Bergantz, 2008). Therefore, the secular evolution of rhyolite compositions not only traces crustal maturation but also encodes information about the prevailing tectonic regime. or are formed through a hybrid process that involves mineral fractionation of mafic to andesitic magmas, either combined with or without the assimilation of felsic components (Ayalew et al., 2002; Bacon and Druitt, 1988; Cameron and Hanson, 1982; Grove et al., 1997).

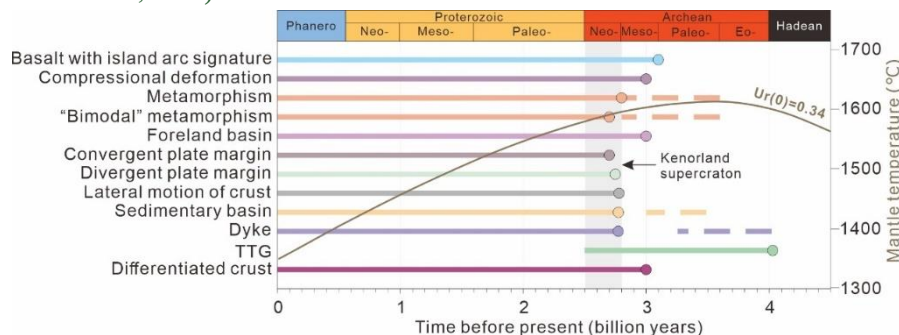


Figure 1: The beginning ages (circles) and age ranges (solid and dashed lines) of proposed major geological archives for plate tectonics. The solid lines correspond to the widespread and continual presence of these geological archives, whereas the dashed lines indicate sporadic occurrences of such geological events. Three ingredients are crucial to understanding the onset of plate subduction: (1) a rigid and more differentiated continental crust; (2) mutual movements of plates; (3) geological evidence for plate margin interaction. The beginning ages and age ranges for a possible rigid continental lithosphere are represented by differentiated crust (Dhuime et al., 2015), tonalite-trondhjemite-granodiorite series (TTGs) (Bowring and Williams, 1999; Moya and Martin, 2012), giant vertical dyke (Ernst et al., 2021) and sedimentary basins (Siahi et al., 2016; Srinivasan and Ojakngas, 1986; Wingate, 1999). The initiation of lithospheric mobility of continents is constrained by paleomagnetic evidence (Cawood et al., 2018). The geological evidence for plate margin interaction can be indicated by the occurrence of divergent (Bradley, 2008; Cawood et al., 2018) and convergent plate margins (Condie, 2021), foreland basin (Burke et al., 1986; Catuneanu, 2001; Corcoran and Mueller, 2007), paired

metamorphic rocks (Brown and Johnson, 2019a; Brown et al., 2020), compressional deformation (Cawood et al., 2018) and basalts with island arc signature (Brown et al., 2020). The thermal model for ambient mantle for Urey (Ur) ratio of 0.34 is also shown (Herzberg et al., 2010).

However, rhyolites associated with differentiation processes of basalts must be relatively minor, as large proportions of rhyolites are rarely differentiated from mafic magmas, even those linked with mafic large igneous provinces (Halder et al., 2021). While large amounts of basalts were presented in the early Archean, significant volumes of rhyolites were not formed through the differentiation of parental mafic magmas before 3.0 Ga (Tang et al., 2016). This cannot be attributed to preservation biases, as rhyolites are more resistant to erosion and more likely to be preserved compared to ultramafic and intermediate rocks. Most rhyolites exhibit high SiO_2 and Al_2O_3 contents and distinct negative Ba, Sr, Eu anomalies, suggesting that they originated at shallow crustal depth within the feldspar stability field (Halder et al., 2021). Therefore, the initial formation of large volumes of highly evolved rhyolites likely depended on the emergence of a more differentiated continental crust on Earth, and thus serve as key archives of crustal maturation. If the earliest large-volume rhyolites shared compositional features similar to those of the Phanerozoic, it would suggest that continental crust had evolved under the influence of modern-style plate tectonics. Here, we test the hypothesis that if the geochemical evolution of rhyolites through Earth's history can reflect the stabilization of rigid lithosphere, enabling plate tectonics. We integrate global rhyolite geochemistry with supercontinent cycles, metamorphic records, and mantle cooling models to resolve this debate.

2 Data and methods

Here we compiled geochemical data for rhyolites spanning 4.0 Ga to present from the EarthChem database (<http://www.earthchem.org/portal>) to investigate the composition and evolution of the continental crust. A dataset of ~21,252 rhyolitic volcanic samples was compiled (Supplementary Table S1, as of 5 November 2022), while another dataset of ~1,208 rhyolites from bimodal volcanic suites was also extracted (Supplementary Table S2). To ensure that only nearly pure rhyolites or high-silicon volcanics were considered, we excluded all geochemical data with $\text{SiO}_2 < 68$ wt%. We analyzed the variation trends of SiO_2 , MgO , and Al_2O_3 over time, as these three oxides are the main components of present-day continental crust. We also examined the initial neodymium isotopes (ϵNd_i) of rhyolites during different ages to identify contributions from juvenile or ancient crusts. Moreover, we plotted the variations of other oxides and trace elements for rhyolitic volcanics, such as Na_2O - K_2O - CaO and Ba, which are mainly dominated by feldspar (as shown in Fig. 2 and supplementary figures). The change in whole-rock $\text{Eu}/\text{Eu}^* [\text{Eu}_N/(\text{Sm}_N * \text{Gd}_N)^{1/2}]$ ratios over time was investigated, which may indicate the depth and compositions of melt source rocks during different time periods. Furthermore, the moving averages of the oxides and trace elements of rhyolites were calculated over time using LOESS regression with weighted bootstrap resampling ($n = 1000$) (Li et al., 2019a). We present our results as means with associated 1σ - and 2σ -standard-error uncertainties for 350-Myr intervals between 0 and 3.5 Gyr in Fig. 2. We also constructed kernel density estimates (KDEs) for each 500-Myr-bin to present the distribution of these

proxies, as well as the medians, inter-quartile range (IQR, 25%–75%) and range within 1.5IQR. The peaks of KDEs and medians show remarkably similar trends and both of them match the moving average curves (Fig. 2).

We also collected representative crustal velocity structure, which constrained by Deep Seismic Sounding method, in Archean Cratons (Musacchio et al., 2004; Kuusisto et al., 2006; Xia et al., 2017; Durrheim and Green, 1992; Drummond and Collins, 1986; Drummond, 1988; Vijaya Rao et al., 2015; Kaila and Sain, 1997) to investigate the seismic features of the Archean craton and the modern global continental crust. Considering the younger magmatic events and sedimentary covers, we assume that the upper crustal velocity is the minimum seismic velocity for these cratons.

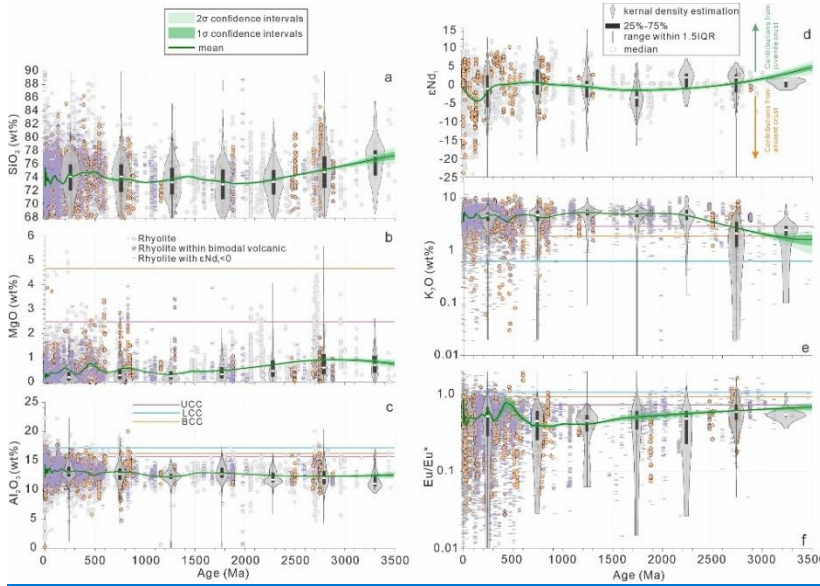


Figure 2: The features of (a)SiO₂; (b) MgO; (c) Al₂O₃, (d) initial neodymium isotopes (εNd), (e) K₂O and (f) Eu/Eu* of rhyolites through time. The compositions of present-day continental crust are adapted from (Rudnick and Gao, 2003). 350-Myr-bin moving average of rhyolite compositions together with the 1σ and 2σ confidence intervals acquired by bootstrap resampling (numbers=1000) are shown. The kernel density estimations, medians, inter-quartile range (IQR, 25%–75%) and range within 1.5IQR of each 500-Myr-bin are also shown for all data without rejection. UCC=upper continental crust; LCC=lower continental crust; BCC=bulk continental crust.

3 Results

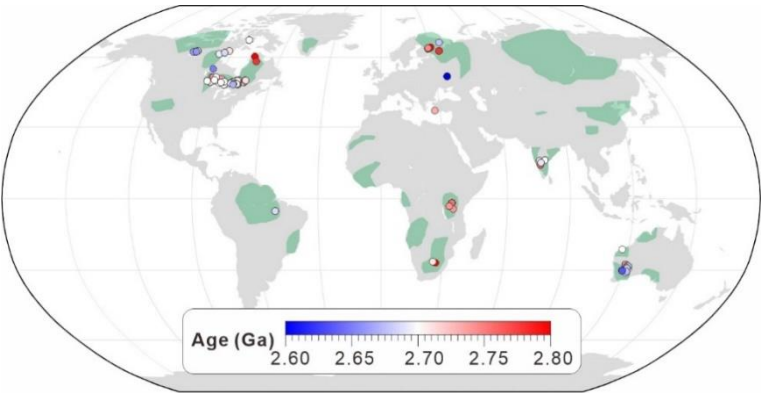
Secular variations in rhyolites show key evolutionary trends culminating in statistically significant diagnostic developments at ca. 2.7 Ga (Fig. 2), characterized by the increased prevalence of rhyolites exhibiting specific compositional features (e.g., elevated K₂O, pronounced negative Eu anomalies) that become more characteristic of post-2.7 Ga and Phanerozoic examples. This change is also characterized by the increased prevalence of rhyolites with expanded compositional ranges (SiO₂, MgO, Al₂O₃, Na₂O, K₂O, CaO, and Ba) particularly for K₂O (IQR increase >40%) and Eu/Eu*, which are comparable to those observed in rhyolites since that time. These ~2.7 Ga rhyolites emerged and were distributed across ancient cratons, rather than

115 being confined to any specific region (Fig. 3). The averages of SiO₂ (Fig. 2a), MgO (Fig. 2b), K₂O (Fig. 2e), and Ba (Fig. S1) all show a distinct shift at ca. 2.7 Ga, while the evolutionary trends of Al₂O₃ (Fig. 2c) and Na₂O (Fig. S1) in rhyolites remain relatively constant throughout Earth's history. Another notable feature at 2.7Ga (Fig. 2d) is the frequent occurrence of rhyolites with negatively initial Nd isotopes (ϵNd_t values between 0 and -10), indicating a significant increase in contributions from ancient and differentiated continental crust.

120 Furthermore, the late Archean craton crust structure (volume percentage: 40% for upper crust, 34% for middle crust, 26% for lower crust) constrained by crustal seismic velocity is similar with that of modern global continental crust (volume percentage: 38% for upper crust, 26% for middle crust, 36% for lower crust) (Fig. 4a). The minimum upper crustal seismic velocity of Archean cratons is negatively correlated with cratonic age, suggesting that younger Archean blocks possess a more felsic crustal composition (Fig. 4b). This trend may reflect the formation of older Archean cratons from a relatively primitive mantle source. After ~2.7 Ga, the upper crustal velocities of these Archean cratons are systematically lower than those of pre-~2.7 Ga cratons, indicating a significant compositional shift in craton formation processes.

125 This These obvious features in rhyolites and crustal structure coincides with a period of maximum rates of continental crustal reworking since 4.5 Ga (Dhuime et al., 2012) (Fig. 4a5a). Throughout the Archean era, a clear trend towards enhanced negative Eu anomalies in rhyolites was observed, with a marked intensification towards the end (Fig. 2f). However, rhyolites from this

130 period have higher SiO₂ (Fig. 2a) and CaO contents (Fig. S1) than those formed from other periods. Furthermore, elevated MgO in Neoproterozoic samples (Fig. 2b) may reflect transitional crustal sources with residual mantle contributions, consistent with hotter Archean geotherms (Herzberg et al., 2010).



135 **Figure 3: The distributions of ~2.7±0.1 Ga rhyolites on Earth. The filled circles represent the location of rhyolite samples. The light green filled areas cover the scope of global cratons. We should note that the significant amount of rhyolite around 2700 million years ago, is not confined to any particular region but instead scattered across ancient cratons.**

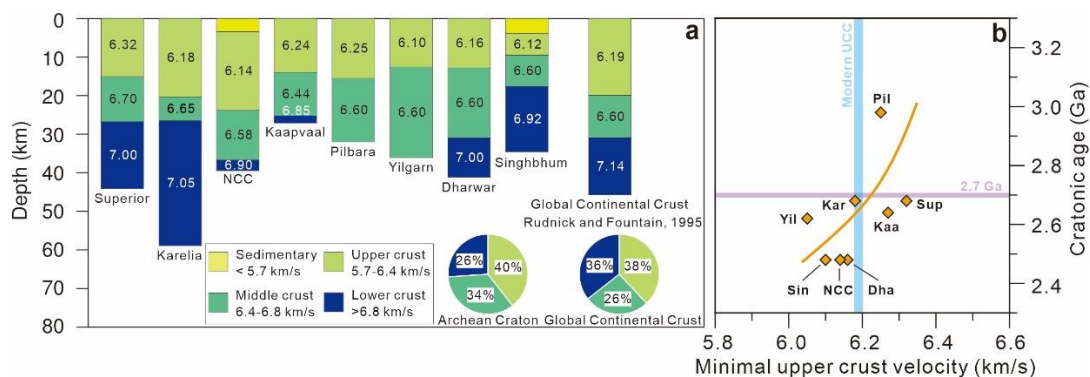


Figure 4: The velocity structure of Archean cratons and global continental crust. (a) Representative cratons' velocity structure (Musacchio et al., 2004; Kuusisto et al., 2006; Xia et al., 2017; Durrheim and Green, 1992; Drummond and Collins, 1986; Drummond, 1988; Vijaya Rao et al., 2015; Kaila and Sain, 1997) and global continental crust velocity structure (Rudnick and Fountain, 1995). (b) Minimal upper crust velocity versus cratonic age, with the fitting curve marked by orange solid line. The light blue line shows the average velocity of modern upper crust (UCC). Abbreviation: Sup: Superior, Kar: Karelia, NCC: North China Craton, Kaa: Kaapvaal, Pil: Pilbara, Yil: Yilgarn, Dha: Dharwar, Sin: Singhbhum.

4 Discussions

4.1 Crustal differentiation and lithospheric rigidization

The compositional transition of continent crust seemingly occurred in the Neoproterozoic. In the early Meso- to Paleoproterozoic, the maturity of the continental crust remained heterogeneous, as evidenced by the compositional variations of rhyolites (Fig. 2). Some rhyolites with high MgO (> 2.5 wt%) (Fig. 2b) and high CaO contents (Fig. S1) could be attributed to a high proportion of mafic crusts and/or detrital minerals, as demonstrated by the less negative Eu anomalies observed in the early Archean rhyolites (Fig. 2f). The emergence of voluminous, silica-rich rhyolites (with their SiO₂, εNd_t values, and K₂O comparable to present-day rhyolites; Figs. 2 and S1) since 2.7 Ga signals pervasive access to evolved felsic sources, as evidenced by enriched Nd isotopes (εNd_t < -2) in >60% of samples post-2.7 Ga, likely driven by continental thickening to ~55 km (Tang et al., 2021).

A key clarification is that our model does not entirely exclude basaltic fractionation as a rhyolite source but emphasizes its minor role before 3.0 Ga. This is supported by the scarcity of evolved rhyolites in pre-3.0 Ga records (Fig. 2) and observations constraints showing that large volumes of rhyolites rarely differentiate from mafic magmas (Halder et al., 2021). Instead, the differentiated crust required for rhyolite anatexis primarily derives from the long-term accumulation of tonalite-trondhjemite-granodiorite (TTG) suites and its weathering products, the dominant components of Archean felsic crust. TTGs formed through multiple interconnected processes: partial melting of hydrated mafic crust at high pressure (Moyen and Martin, 2012; Sotiriou et al., 2023), amphibole/plagioclase-dominated fractional crystallization from basaltic/dioritic magmas (Liou and Guo, 2019; Laurent et al., 2020; Smithies et al., 2019), and hybridization of melts from sinking greenstone belts (Huang et al., 2025).

Critically, these processes operated at moderate pressures (<1.5 GPa; Smithies et al., 2019), with hydration sourced from primordial mantle water or seawater-hydrothermal alteration of oceanic crust/plateau (Ma et al., 2025; Smithies et al., 2021; Xiong et al., 2025; Zhang et al., 2025; Zhao et al.2025), generating a compositionally heterogeneous felsic reservoir.

165 The temporal build-up of TTGs was probably central to rhyolite petrogenesis. From 3.5 to 2.7 Ga, TTGs accumulated progressively (Fig. 1; Cawood et al., 2018; Sun et al., 2021), reaching a critical volume by ~2.7 Ga sufficient to serve as a source for large-scale rhyolitic melts. This timeline aligns with the surge in evolved rhyolites, as TTGs provided a mature, differentiated crust susceptible to anatexis. We note that the extraction of rhyolitic melts from TTG-dominated crust is consistent with the “mush model” of Bachmann and Bergantz (2008), wherein interstitial melts are segregated from crystalline
170 mushes at intermediate crystallinities. The post-2.7 Ga rhyolites, with elevated K₂O and pronounced negative Eu anomalies, reflect melting within a feldspar-stable, shallow crustal environment—a signature akin to the “cold-wet-oxidized” rhyolites of Phanerozoic subduction zones (Bachmann & Bergantz, 2008). This implies that by ~2.7 Ga, the crust had not only matured compositionally but also stabilized mechanically, allowing for the development of large, coherent mush zones from which melt could be efficiently extracted.

175 Isotopic evidence reinforces this connection. Smithies et al. (2021) documented a secular shift in TTG oxygen isotopes from mantle-like ($\delta^{18}\text{O} \sim 5.3\text{‰}$) to crustal values (>6‰) by 2.7 Ga, paralleling the isotopic enrichment in coeval rhyolites (Fig. 2d). Furthermore, the ~2.7 Ga shift from TTG-dominated to K-rich granitoid magmatism (Laurent et al., 2014), together with our rhyolite data, reflects a feedback loop: (1) TTG-driven crustal growth leads to continental thickening, and (2) radiogenic heat from thickened crust sustained prolonged melting, ensuring the large volumes of evolved rhyolites observed after ~2.7 Ga.
180 This crustal maturation is further corroborated by the systematic decrease in minimum upper crustal seismic velocities of Archean cratons post-2.7 Ga (Fig. 4b), indicating a more felsic and differentiated crustal composition, consistent with the geochemical trends observed in rhyolites (Fig. 5c).

This thickening also promoted large-scale continental emergence above sea level (Pons et al., 2013) and amplified erosional efficiency (Lee, 2014), increasing the flux of felsic detritus-derived from TTGs to marine systems. A sharp rise in seawater
185 ⁸⁷Sr/⁸⁶Sr (Shields, 2007) (Fig. 4a5a, b) corroborates intensified TTGs-dominated continental crust weathering, while burial of mature sediments facilitated crustal melting, further driving crustal differentiation and silica-rich rhyolites production. That is the reason why large volumes of rhyolites have not been present on Earth until just 2.7 Ga (Fig. 2), when mature sedimentary packages began to occur, starting from the late Mesoarchean (Reimink and Smye, 2024). This crustal reworking process is further amplified when TTGs themselves are remelted: as older TTGs (with mantle-like isotopes) are melted and mixed with
190 younger, reworked TTGs (Smithies et al., 2021), the resulting rhyolites inherit both the fractionation signatures (Eu/Eu*) and isotopic enrichment ($\epsilon\text{Nd}_t < -2$) observed in our data. By ~2.7 Ga, the cumulative continental volume reached ~75% of its modern size (Fig. 4b5b) (Dhuime et al., 2012). Systematic increases in K₂O (Fig. 2e) and negative Eu anomalies (Fig. 2f) of rhyolites, alongside declining Na₂O/K₂O and increasing ⁸⁷Sr/⁸⁶Sr ratios of global intermediate to felsic rocks (Keller and

Schoene, 2012), underscore a transition from mafic to felsic upper crust at that time. This is further supported by sedimentary
proxies (e.g., Cr/U, Ni/Co-MgO) (Smit and Mezger, 2017; Tang et al., 2016) and $\delta^{51}\text{V}$ data indicating felsic dominance post-
3.0 Ga (Tian et al., 2023). The increase in negative Eu anomalies of rhyolites from the Archean to Phanerozoic is thought to
be related to a secular evolution in continental crust (Fig. 2f), indicating crustal maturation, which is corroborated by the peak
crustal reworking rates at ~2.7 Ga (Fig. 4a5a).

In essence, the ~2.7 Ga rhyolite transition marks the culmination of a protracted process: TTGs, formed through diverse
mechanisms and accumulated over hundreds of millions of years, built a differentiated crust. As this crust matured to a
threshold which is not arbitrary but reflects the convergence of multiple processes: (1) TTG volume reached a critical mass to
sustain widespread melting; (2) crustal thickness stabilized feldspar-rich residues, amplifying Eu anomalies; (3) and mantle
cooling reduced magmatic perturbations, allowing the crust to retain the differentiated signature necessary for rhyolite
production. These signatures align with the secular shift from tonalite-trondhjemite-granodiorite (TTG) suites to K-rich
granites, as documented in global cratons (Cawood et al., 2018; Laurent et al., 2014), though the presence of the oldest felsic
crust (Acasta Gneiss Complex) can be traced back to ca. 4.0 billion years ago (Bowring and Williams, 1999). It also appears
that 60 to 90% of the volume of continental crust formed during 3.5–2.5 Ga (Cawood et al., 2022; Dhuime et al., 2012;
Hawkesworth et al., 2017). Such compositional evolution implies enhanced crustal reworking and stabilization, driven by
thickening of the lithosphere (Tang et al., 2021) and the accumulation of radioactive heat-generating elements (e.g., K, U, Th).
The emergence of peraluminous granites (Zhu et al., 2022) and widespread giant dyke swarms post-2.7 Ga (Ernst et al., 2021)
reflect lithospheric stabilization, enabled by secular mantle cooling (Fig. 1) (Herzberg et al., 2010). Then, reduced melt
impregnation enhanced lithospheric viscosity (Sizova et al., 2010), favoring stress focusing at craton margins (Rey et al., 2014)
and replacing the early Archean vertical tectonics (i.e., dome-and-keel) with mechanisms dominated by horizontal thrust belts
(Van Kranendonk, 2004).

4.2 Plate margin activation and supercraton assembly

The ~2.7 Ga crustal stabilization, rooted in TTG accumulation and lithospheric rigidization, marked a threshold in lithospheric
strength that enabled the activation of interconnected plate margin processes. The temporal concurrence of (1) rhyolite
geochemical shifts (K_2O , Eu/Eu^*), (2) peak metamorphism (Fig. 4e), and (3) passive margin emergence (Fig. 4f5f) at ~2.7 Ga
(Figs. 2, 4f5f and S1) is temporally congruent with the earliest definitive evidence of interconnected plate boundary processes.

This transition bridges the crustal differentiation documented in rhyolites (section 4.1) with the emergence of tectonic features diagnostic of plate tectonics, linking deep crustal maturation to surface dynamics.

The mechanical coherence required for plate-scale motion, facilitated by thickened, rigid continental lithosphere, coincides with the first definitive records of plate boundary assemblages. Passive margins (Bradley, 2008) and rift-to-drift transitions (Cawood et al., 2018) signal the onset of seafloor spreading, while foreland basins such as the Witwatersrand (Burke et al., 1986) record crustal loading from convergent margins. These features, alongside bimodal volcanics (Fig. 4f5f), mirror Phanerozoic tectonic archives and align temporally with the rhyolite geochemical shift (K_2O , Eu/Eu^* ; Fig. 2). Critically, the hydrous conditions required for TTG formation, linked to seawater-hydrothermal alteration of oceanic crust (Xiong et al., 2025; Zhang et al., 2025), would have persisted at these nascent plate margins, sustaining the crustal reworking that fed rhyolite anatexis. The compressional deformation and basalts with island arc signature (Brown et al., 2020) imply subduction initiation occurred since 3.0 Ga (Fig. 1), albeit localized, prior to sustained plate tectonics. This kind of consideration is further supported by the occurrence of a large number of paired high T/P and intermediate T/P features (Fig. 4e5e, f) during the Neoarchean (Brown and Johnson, 2019a; Brown et al., 2020). This framework reconciles disparate proxies, from rhyolite geochemistry to metamorphic belts, and establishes ~2.7 Ga as a pivotal milestone in Earth's shift to modern geodynamics. The synchronicity of rhyolite geochemical shifts (Fig. 5c), crustal reworking rates (Fig. 5a), metamorphic peaks (Fig. 5e), and the stabilization of felsic upper crust as evidenced by seismic velocities (Fig. 4) underscores a fundamental reorganization of Earth's geodynamic regime at ~2.7 Ga.

The rigidity of differentiated crust enabled stress transmission at 2.7 Ga, facilitating horizontal motion and A-type granite emplacement (Fig. 4d5d) in compressional settings (Condie et al., 2023). Although ophiolites became widespread post-900 Ma (Stern, 2005), the oldest example (Kusky et al., 2020) and subduction-related records (Condie and Stern, 2023) suggest that subduction likely initiated on a local scale by at least 2700 million years ago. Furthermore, paleomagnetic data from the Kaapvaal, Pilbara, and Superior cratons reveal significant relative motion between 2.7 and 2.4 Ga (Cawood et al., 2018). For instance, the Kaapvaal and Superior cratons show >5,000 km of displacement along small-circle trajectories. The culmination of horizontal movements in continental crusts is exemplified by supercontinent assembly, and there is evidence indicating that the earliest supercontinent may have formed at 2.7 Ga. Here, the assembly of Kenorland, the earliest supercraton, at ~2.7 Ga (Bleeker, 2003) provides a key tectonic context. Supercontinent assembly demands large-scale horizontal motion, which is only feasible with rigid, buoyant crust, properties conferred by the cumulative TTG-derived felsic reservoir (Laurent et al., 2020; Sun et al., 2021). By 2.7 Ga, TTG volumes had reached a critical threshold (Sun et al., 2021), creating a buoyant framework that resisted subduction and enabled cratonic collisions. This assembly coincided with peak metamorphism (Fig. 4e5e) and the stabilization of thick (~55 km) lithosphere (Tang et al., 2021), underscoring how TTG-driven crustal growth laid the mechanical foundation for supercraton formation. Rhyolites, as sensitive tracers of crustal maturity, capture this process:

their enriched Nd isotopes ($\epsilon\text{Nd}_t < -2$) and evolved geochemistry reflect the reworking of ancient TTG crust during collisional events, linking supercontinent assembly to enhanced crustal recycling.

Our results also align with the 'stagnant-lid to plate tectonics transition' model (Cawood et al., 2018), which posits that Neoarchean lithospheric strengthening (driven by mantle cooling and crustal thickening) enabled stress transmission necessary for plate-scale motion. However, whereas previous studies primarily invoked passive margins and foreland basins as evidence (Bradley, 2008; Burke et al., 1986), this study provides direct geochemical proxies (K_2O , Eu/Eu^*) for deep crustal maturation that temporally couples with these plate boundary features. The synchronicity of enriched Nd isotopes in rhyolites (Fig. 2d) with peak crustal reworking rates (Fig. 4a5a) further constrains the timing of differentiated continental emergence—a prerequisite for generating felsic sedimentary archives used in other models. The temporal linkage between crustal maturation, supercraton assembly, and the oldest subduction records resolves longstanding debates on the prerequisites for sustained plate tectonics.

4.3 Reconciling conflicting models for Archean geodynamics

The ~ 2.7 Ga transition documented in rhyolitic geochemistry provides a pivotal framework to resolve long-standing debates between proponents of "early" (>3.0 Ga) and "late" (<1.0 Ga) onset models for plate tectonics. This transition, rooted in the coevolution of TTG suites and rhyolites, reconciles disparate evidence by linking crustal maturation to the emergence of sustained plate dynamics, without requiring abrupt tectonic shifts.

Proponents of early subduction cite sporadic occurrences of arc-like basalts (e.g., $\text{Th}/\text{Nb} > 0.2$) in >3.0 Ga greenstone belts (Polat et al., 2002) and Hadean zircon signatures (Turner et al., 2020), with the latter suggesting andesitic sources akin to modern subduction settings. While advocates of late onset emphasizes the scarcity of definitive ophiolites and ultrahigh-pressure metamorphism before 1.0 Ga (Stern, 2005). Although these lines of evidence are compelling for their respective models, both frameworks overlook a key prerequisite: the crustal maturity required for plate-scale motion.

For early subduction models, Zhang et al. (2023) showed that 4.0 Ga Acasta gneisses preserve Si-O isotopic signatures indicating minimal supracrustal recycling, while widespread surface material reworking emerged only after 3.8 Ga. Our geochemical model, which attributes the ~ 2.7 Ga shift to the maturation of TTG-dominated crust, provides a framework to reconcile these conflicting hypotheses. This reconciliation is further strengthened by recent insights into the petrogenetic mechanisms of early felsic crust. Notably, Ma et al. (2025) demonstrates that Fe and Zn isotopes in Neoarchean plagiogranites, analogous to Archean TTGs, record H_2O -saturated melting of altered oceanic crust (AOC) at shallow depths (2–6 kbar) under high thermal gradients (1200–4500 $^\circ\text{C}/\text{GPa}$), consistent with a mantle plume setting. Their findings, coupled with the composition of the Hadean Idiwhaa gneisses (Aarons et al., 2020), confirm that the production of evolved, felsic crust, a

prerequisite for rigidity, can occur through mechanisms other than modern-style subduction, such as plume-induced melting of hydrated mafic proto-crust.

Furthermore, the purported pre-3.0 Ga plate boundary signatures (e.g., boninitic magma in Isua, Greenland) lack synchronous evidence for large-scale lithospheric mobility. Paleomagnetic data show no significant relative motions between cratons prior to 2.8 Ga (Cawood et al., 2018), and passive margins—key indicators of seafloor spreading, are absent before 3.0 Ga (Bradley, 2008; Fig. 4f5f). Crucially, the limited compositional range of pre-2.7 Ga rhyolites (e.g., low K₂O, weak Eu anomalies; Fig. 2e, f) reflects immature crust incapable of sustaining rigid plate behavior. This implies that even if localized “subduction” occurred, it operated within a non-plate tectonic regime dominated by vertical “dome-and-keel” dynamics (Van Kranendonk, 2004), where high mantle temperatures (~100–150°C hotter than present) prevented stress transmission across lithospheric scales (Sizova et al., 2010).

Here, the classification of rhyolites by Bachmann and Bergantz (2008) into “hot-dry-reduced” and “cold-wet-oxidized” end-members provides a useful lens. Pre-2.7 Ga rhyolites, with their weaker Eu anomalies and lower K₂O, resemble the “hot-dry-reduced” type, consistent with a non-subduction, plume-influenced setting. In contrast, the post-2.7 Ga surge in rhyolites with strong Eu anomalies and high K₂O aligns with the “cold-wet-oxidized” signature, indicative of melting in a hydrated, feldspar-rich crust under conditions akin to modern subduction zones. This geochemical shift thus marks not only crustal maturation but also a fundamental change in the tectonic environment—from a vertically dominated, “stagnant-lid” regime to one capable of sustaining horizontal plate motions.

Conversely, late-onset models overlook the diagnostic shifts in Neoarchean archives. The surge in rhyolite K₂O (3–5 wt%) and negative Eu anomalies ($\text{Eu}/\text{Eu}^* = 0.3\text{--}0.6$) at ~2.7 Ga (Fig. 2e, f) coincides with: (1) the oldest foreland basins (e.g., Witwatersrand; Burke et al., 1986), (2) paired intermediate/high T/P metamorphic belts (Brown & Johnson, 2019b; Fig. 4e5e), and (3) paleomagnetically constrained cratonic displacements (>5,000 km for Superior-Kaapvaal; Cawood et al., 2018). Furthermore, Zhong et al. (2025) documented an inverted metamorphic gradient in the North China craton (NCC) Zhanhuang thrust system, where peak metamorphic conditions dropped from 11 kbar/700°C to 4 kbar/500°C over 1 km, indicating large-scale thrust stacking at ~2.5 Ga. This structural evidence for horizontal motion coincides with our rhyolite geochemical shift, as both record rigid lithospheric behavior. Similarly, Zhong et al. (2021) identified Alpine-style nappes in the NCC with ~3560 km of horizontal transport between 2.7 and 2.5 Ga, directly demonstrating the capacity for plate-scale motion by the late Archean. These features collectively signify horizontal motions and convergent margins, hallmarks of plate tectonics absent

in pre-3.0 Ga records. The rise of peraluminous granites and giant dyke swarms post-2.7 Ga (Ernst et al., 2021) also reflects stabilized lithosphere (Fig. 1). Cawood et al. (2023) proposed Earth's tectonic evolution progressed through three modes: stagnant lid, squishy lid (distributed deformation), and rigid plate tectonics, with the latter emerging in the late Archean. Our rhyolite records align with this transition: the 2.7 Ga shift in K_2O , Eu/Eu^* , and ϵNd_t coincides with rigid continental lithosphere stabilization, a hallmark of the squishy-lid to plate tectonics transition. This is corroborated by synchronous peak crustal reworking (Dhuime et al., 2012; Fig. 4a5a) and Kenorland assembly (Bleeker, 2003).

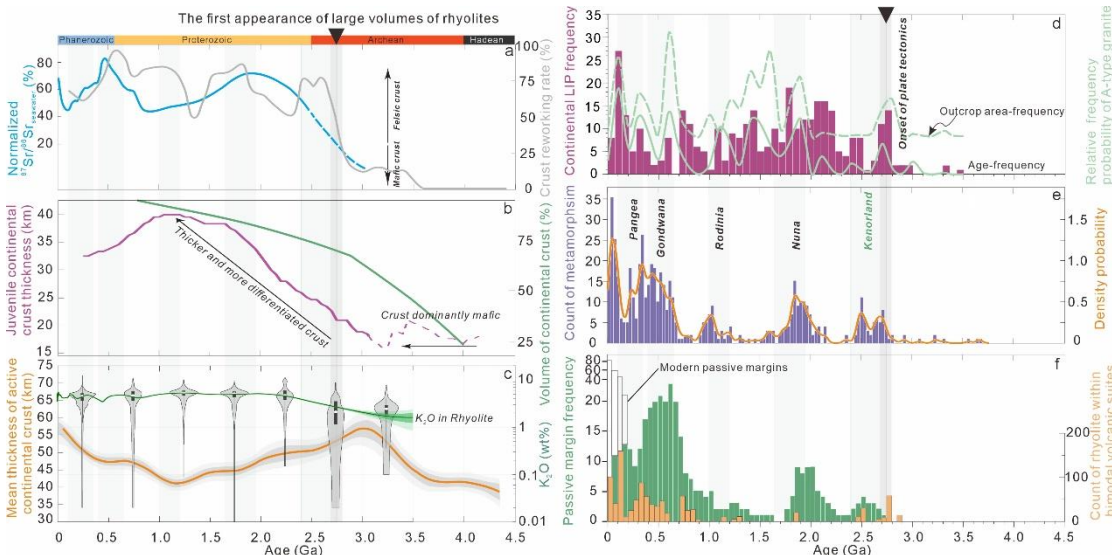


Figure 45: Secular changes of (a) Normalized seawater $^{87}Sr/^{86}Sr$ curve (Shields, 2007) and the degree of reworking of the continental crust (Dhuime et al., 2012); (b) Continental growth curve (Dhuime et al., 2012) and thickness of juvenile continental crust (Dhuime et al., 2015); (c) K_2O in global rhyolites and mean thickness of active continental crust (Tang et al., 2021); (d) the numbers of continental large igneous provinces (LIPs) (Ernst et al., 2021), and the relative age/outcrop area-frequency probability of A-type granites (Condie et al., 2023) through Earth's history; (e) Ages and probability density function for global metamorphism (Brown and Johnson, 2019b); and (f) the distributions of passive margins and the count of rhyolites within bimodal volcanics (Bradley, 2008). Note the convergence of multiple proxies (rhyolite K_2O , crustal reworking, metamorphic peaks, etc.) within the 2.7 Ga transition window.

Here, the global rhyolite dataset provides quantifiable proxies for crustal maturation, complementing structural and metamorphic archives. Widespread post-2.7 Ga rhyolites with $K_2O > 3$ wt% and $Eu/Eu^* < 0.6$ reflect continental lithosphere thickening to ~55 km (Tang et al., 2021), a process tied to "modern-style" rigidity. This is uniquely recorded in rhyolites as their geochemistry requires melting of evolved continental sources, a process contingent on differentiated crustal reworking. This rigidity enabled stress transmission across craton margins (Rey et al., 2014), replacing vertical dome-and-keel tectonics with horizontal thrusting. Huang et al. (2022) documented coexisting divergent (MORB-like) and convergent (arc-like)

assemblages in the NCC at 2.55–2.51 Ga, providing direct evidence for linked plate boundaries—an essential feature of our ~2.7 Ga transition model.

330 Notably, the ~2.7 Ga rhyolite shift predates the 2.5 Ga structural records (Zhong et al., 2021, 2025) by ~200 Myr, suggesting crustal rigidization (recorded by rhyolites) was a prerequisite for large-scale plate motion. This sequence of crustal maturation, lithospheric strength, and plate interaction, resolves the "early vs. late" debate by showing localized subduction (e.g., 3.0 Ga arc basalts; Brown et al., 2020) operated within a non-plate regime until cumulative mantle cooling and crustal stabilization enabled global connectivity. This timing aligns with the "Juvenile Earth" phase (3.2–2.5 Ga) (Cawood et al., 2023), where
335 craton stabilization laid the groundwork for interconnected plate dynamics. By linking rhyolite geochemistry to these transitions, our study bridges crustal processes and tectonic mode shifts, providing a new constraint on Earth's transition to plate tectonics. The integration of seismic velocity structure (Fig. 4) with geochemical and metamorphic proxies (Fig. 5) provides a multi-proxy validation of the ~2.7 Ga transition, highlighting the coevolution of crustal composition, rigidity, and plate boundary processes.

340 5 Conclusions

The ~2.7 Ga geochemical shift in rhyolites, marked by elevated potassium, negative Eu anomalies, and enriched Nd isotopes, signifies a lithospheric strength threshold enabling plate-scale stress transmission. This transition is rooted in the protracted accumulation and maturation of TTG suites, which built the differentiated, rigid continental crust required for sustained plate tectonics.

345 Critically, this crustal rigidization is independently recorded by the seismic architecture of Archean cratons. The systematic decrease in minimum upper crustal velocities post-~2.7 Ga provides physical evidence for a more felsic and differentiated crustal composition, aligning temporally with the geochemical maturation documented by rhyolites. Our model therefore reconciles conflicting hypotheses by demonstrating that while localized subduction may have occurred earlier, sustained, globally connected plate tectonics emerged only after ~2.7 Ga. This shift was enabled when cumulative mantle cooling and
350 crustal differentiation stabilized a rigid lithosphere, as evidenced by the synchronicity of rhyolite geochemistry, peak crustal reworking rates, metamorphic patterns, and diagnostic changes in crustal seismic structure.

The global rhyolite record, integrated with geophysical and structural proxies, uniquely captures this transition. It thereby provides a definitive geochemical link between deep crustal processes (e.g., crustal reworking) and the surface tectonic manifestations of supercraton assembly and plate margin activation. This multi-proxy framework highlights the fundamental
355 role of crustal maturation in Earth's shift to modern geodynamics, with implications for understanding the tectonic evolution of rocky planets.~~Our model reconciles conflicting hypotheses by demonstrating that while localized subduction may have occurred earlier, sustained plate tectonics emerged only after 2.7 Ga, when cumulative mantle cooling and crustal differentiation stabilized rigid lithosphere. The global rhyolite record uniquely captures this transition, thereby providing a~~

geochemical link between deep crustal processes (e.g., crustal reworking) and surface tectonic manifestations (e.g., supercraton assembly). This framework highlights the role of crustal maturation in Earth's shift to modern geodynamics, with implications for linking early planetary evolution to the onset of plate tectonics.

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Data Availability Statement

The data supporting the conclusions can be obtained from the Additional Supporting Information, which are available at <http://www.earthchem.org/portal>.

Author contributions

~~Xiangdong X.D.~~~~Su~~: Conceptualization, Data curation, Investigation, Methodology, Writing - original draft, Writing - review & editing. ~~G. aochun C.-W.~~~~ang~~: [Data curation, Investigation, Methodology, Writing - original draft, Writing - review & editing.](#) ~~Yanjie Y.J.~~~~T.~~~~ang~~: Writing - review & editing. ~~P.~~~~eng~~~~P.~~~~eng~~: Conceptualization, Writing - review & editing. ~~P.L.~~~~:~~~~Writing - review & editing.~~

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