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# Europium anomalies in detrital zircons record major transitions in Earth geodynamics at 2.5 Ga and 0.9 Ga

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## ABSTRACT

**Trace elements in zircon are a promising proxy with which to quantitatively study Earth's long-term lithospheric processes and its geodynamic regimes. The zircon Eu anomaly reflects the crystallization environment of its felsic or intermediate parental magma. In particular, it provides insight into the water content, magmatic redox conditions, and the extent of plagioclase fractionation in the source rock or its occurrence as a cogenetic crystallizing phase from the magma. We performed a statistical analysis of Eu anomalies from a compilation of detrital zircons over geologic time and found a major decrease in Eu anomaly ca. 2.5 Ga and an important increase ca. 0.9 Ga. Coupled with thermodynamic modeling, we suggest that these variations could be due to long-term change in the chemical system of the mafic source from which the intermediate to felsic melt and derived zircons were produced. The 2.5 Ga drop was likely associated with an enrichment in incompatible elements in the mafic source, which extended the pressure-temperature field of plagioclase stability as a cogenetic melt phase. We interpret the 0.9 Ga rise to record increasing hydration of magmagenetic sites due to the general development of cold subduction systems, which would delay and/or suppress the saturation of plagioclase in hydrous magmagenetic sites.**

## INTRODUCTION

The application of large, global geochemical data sets is a valuable approach to qualitatively and quantitatively reconstruct Earth's long-term crustal processes (e.g., Dhuime et al., 2015; Profeta et al., 2015; Luffi and Ducea, 2022). Zircon represents a remarkable and ubiquitous crustal archive, surviving most metamorphic, hydrothermal, weathering, and erosional processes. Thus, zircon provides a record of igneous and/or medium- to high-grade metamorphic processes, avoiding biases associated with whole-rock geochemistry (Szymanowski et al., 2020). Coupled U-Pb dating and trace-element analysis of detrital zircons have been used as a proxy for provenance discrimination and for inferring protolith composition (Belousova et al., 2002; Sundell et al., 2022) and, more recently, for investigating the evolution of petrogenetic processes through time (e.g., Balica et al., 2020; Jara et al., 2021; Tang et al., 2021b; Carrapa et al., 2022a).

Trace-element concentrations in zircons are particularly useful because they reflect the crystallization environment of their parent magma, the occurrence of cogenetic crystallizing mineral phases, temperature conditions, and volatile contents (Szymanowski et al., 2017). However, the study of trace elements in zircons has limitations when applied to detrital populations, which can lead to misinterpretations. For example, the use of the Ti-in-zircon thermometer requires Ti saturation in the magmatic liquid, which cannot be confirmed in detrital records (Fu et al., 2008). The Ce positive anomaly in zircon is sensitive to magmatic oxidation state (Trail et al., 2012), water content, temperature, melt composition (Smythe and Brenan, 2016), several cogenetic mineral phases, and magma cooling conditions (Loader et al., 2022). However, the very low light rare earth element (LREE) content in zircon complicates interlaboratory reproducibility of Ce* estimates. In zircon, the Eu content generally shows a negative anomaly (noted Eu/Eu*; Fig. 1). This is interpreted as the result of preferential incorporation of $Eu^{2+}$ by plagioclase crystallization, leading to Eu depletion in the melt prior to or during zircon saturation (Karakas et al., 2019; Tang et al., 2021b). However, lower Eu content has also been correlated with low water content and/or reducing magmatic conditions, where $Eu^{2+}$ does not substitute into the zircon lattice (Trail et al., 2012; Smythe and Brenan, 2016). Recently, several authors (e.g., Tang et al., 2021a) have proposed using Eu anomalies in zircon as a proxy for crustal thickness. Plagioclase breakdown and garnet stability at high-pressure conditions may oxidize redox-sensitive trace elements, such as Eu, and ultimately affect their incorporation in zircon. However, the Eu anomaly proxy yielded unrealistic values for paleocrustal thickness in studies in the Andes (Carrapa et al., 2022a, 2022b).

Our study focused on the evolution of the Eu anomaly in zircon through time because Eu (like Sm and Gd) is precisely quantified in zircon data sets (Balica et al., 2020). The Eu anomaly has been demonstrated as an excellent proxy with which to track variations in crustal thickness in regional, "modern," and relatively short-lived magmatic systems (Karakas et al., 2019; Tang et al., 2021b; Sundell et al., 2022). Nevertheless, caution should be exercised when applying it to Earth's long-term crustal evolution without considering other key magmagenetic processes,

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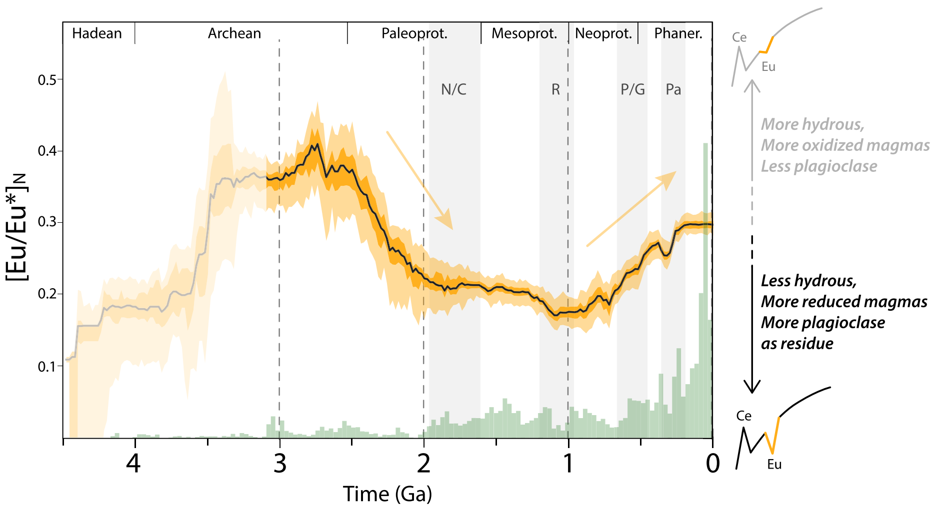


**Figure 1. Zircon Eu anomaly time series using the maximum likelihood of median values distribution from bootstrap iterations applied on a 250 m.y. rolling window at 20 m.y. steps. Orange areas are 1σ and 3σ standard deviations around the average line. Light green histogram shows the frequency of zircon Eu data. Supercontinents: N/C—Nuna/Columbia, R—Rodinia, P/G—Pannotia/Gondwana, Pa—Pangea.**

including the composition of the source, the nature and proportion of cogenetic phases, the influence of water in the system, and the magmatic oxidation state.

We explored the role of these parameters by performing a statistical analysis of global zircon Eu geochemistry from the Mesoarchean (ca. 3.2 Ga) to the present and used the results to discuss how global tectonics varied over time in light of petrologic modeling of average mafic compositions. Using this approach, we tested the role of mafic protolith composition, depth of magmagenesis (i.e., crustal thickness and pressure conditions), and water content controlling the Eu anomaly in zircons. This allowed us to identify two major changes in Earth's lithospheric history at 2.5 Ga and 0.9 Ga.

## DATA AND RESULTS

Detrital zircon U-Pb and trace-element compositions were compiled from 103 rock samples from various continents and cratons. We filtered out metamorphic zircons based on zircon Th/U ratios ($n$ = 6305 data after filtering; Rubatto, 2017; see Item S1 in the Supplemental Material[1]). These zircons gave concordant U-Pb ages and were assumed to have formed in magmatic or high-grade granulitic conditions, derived from intermediate to felsic magmas. The time series was built using a bootstrap analysis by extracting median values with a 500 m.y. rolling window (see Table S1 and Items S1 and S3 in the Supplemental Material for sample locations, single-grain data, and relevant Python codes). The evolution of Eu anomalies (Eu/Eu*) through time showed higher values (~0.37) from 3.5 to 2.5 Ga followed by a significant decrease from 2.5 to 2.0 Ga (Fig. 1). The signal in Eu anomaly appeared to be relatively stable at lower values (~0.19), forming a plateau, from 2.0 to 0.9 Ga. The Eu anomaly then increased from 0.9 Ga to present (up to 0.29). For ages older than the Mesoarchean (>3.2 Ga), the trend in Figure 1 is not considered to be statistically supported by enough zircon data (see Item S1).

## DISCUSSION

### The 2.5 Ga Eu/Eu* Decrease Is Not Related to Global Crustal Thickness

From 2.5 Ga to 2.0 Ga, there was a consistent decrease in zircon Eu anomaly (Fig. 1). The onset of decreasing global zircon Eu anomaly broadly coincides with the Archean-Proterozoic transition, which represents a turning point in Earth's tectonic evolution and profound changes in its atmospheric conditions (Hawkesworth et al., 2017). During this time, the rates of banded iron formation (BIF) deposition slowed, and sulfides in sediments became less abundant. This is consistent with the occurrence of a major oxygenation episode in Earth's atmosphere (Gaucher and Frei, 2018). The 2.5 Ga transition was also synchronous with a major discontinuity in the whole-rock geochemical archive (Keller and Schoene, 2012). In a global compilation, mafic rocks (43–51 wt% $SiO_2$) show decreasing abundances of compatible elements (Ni, Cr, and Mg) and increasing abundances of incompatible elements (Na, K, and $[La/Yb]_N$) after the Archean-Proterozoic boundary. These trends have been interpreted as reflecting a decrease in mantle potential temperature through time, which means that higher percentages of melt were extracted during the Archean compared to the Proterozoic. These changes in the amount of mafic material may have modified primary melt compositions and the crustal structure, as the stability of crystallizing phases would be affected.

The zircon Eu anomaly negatively correlates with the evolution of Rb/Sr in juvenile continental rocks. Based on the Rb/Sr record, Dhuime et al. (2015) suggested that crustal rocks became more differentiated after 3.0 Ga, although preserved pre–3.0 Ga continental crust is decidedly dominated by evolved/felsic tonalite-trondhjemite-granodiorite (TTG) suites. Dhuime et al. (2015) also proposed that this record could mark an increase in global crustal thickness from ca. 3.0 Ga to ca. 1.0 Ga. This change in global crustal thickness contradicts the estimates from Tang et al. (2021b) based on the evolution of the zircon Eu anomaly. However, these studies applied empirical relationships between crustal thickness and chemical proxies found for modern-day plate settings to Precambrian lithologies, which may have formed in a different geodynamic regime.

Because most zircons are found in intermediate to felsic rocks, we used an averaged mafic crust composition (Keller and Schoene, 2012) to model partial melting and/or fractional crystallization and inspect cogenetic phase stability at 3.0 and 2.0 Ga. The 3.0 Ga average mafic composition used in Figure 2A is similar to the composition of an enriched Archean tholeiite (Condie, 1981). Such a composition is considered to represent a potential mafic protolith for Archean evolved melt like TTGs (Hernández-Montenegro et al., 2021). The modeled 3.0 Ga pressure-temperature ($P$-$T$) phase diagram (Fig. 2A) shows that the locus of partial melting (i.e., solidus) occurs at ~700–750 °C. The plagioclase mode is strongly dependent on the pressure conditions. Its stability field is below 9 kbar at 950 °C as the garnet-in line starts above 8.5 kbar at 900 °C. Considering two end members of geothermal gradients for the Precambrian crust (i.e., $dT/dP$ of 100 °C/kbar and $dT/dP$ of 50 °C/kbar, respectively) and two conditions of melt loss events (MLE-a at 7 vol% melt and MLE-b at 20 vol% melt; Bowman et al., 2021), no plagioclase is in equilibrium with felsic melt.

With the 2.0 Ga mafic composition, the garnet-in line is equivalent to that of the 3.0 Ga diagram for suprasolidus conditions, but it occurs at higher pressure conditions (~14 kbar) in the subsolidus region. On the other hand, the plagioclase stability field is extended compared to the previous $P$-$T$ phase diagram, with

[1]Supplemental Material. Additional figures, datasets with a compilation of detrital zircon data (U-Pb and trace element) and references; the detailed statistical treatment for zircon U-Pb and trace composition as well as the parameters and solid solution models used for thermodynamic modelling; and links to a GitHub repository that contains two Python codes built to perform bootstrap analysis and timeseries plotting on the zircon U-Pb and trace elements dataset. Please visit https://doi.org/10.1130/GEOL.S.21520947 to access the supplemental material and contact editing@geosociety.org with any questions.

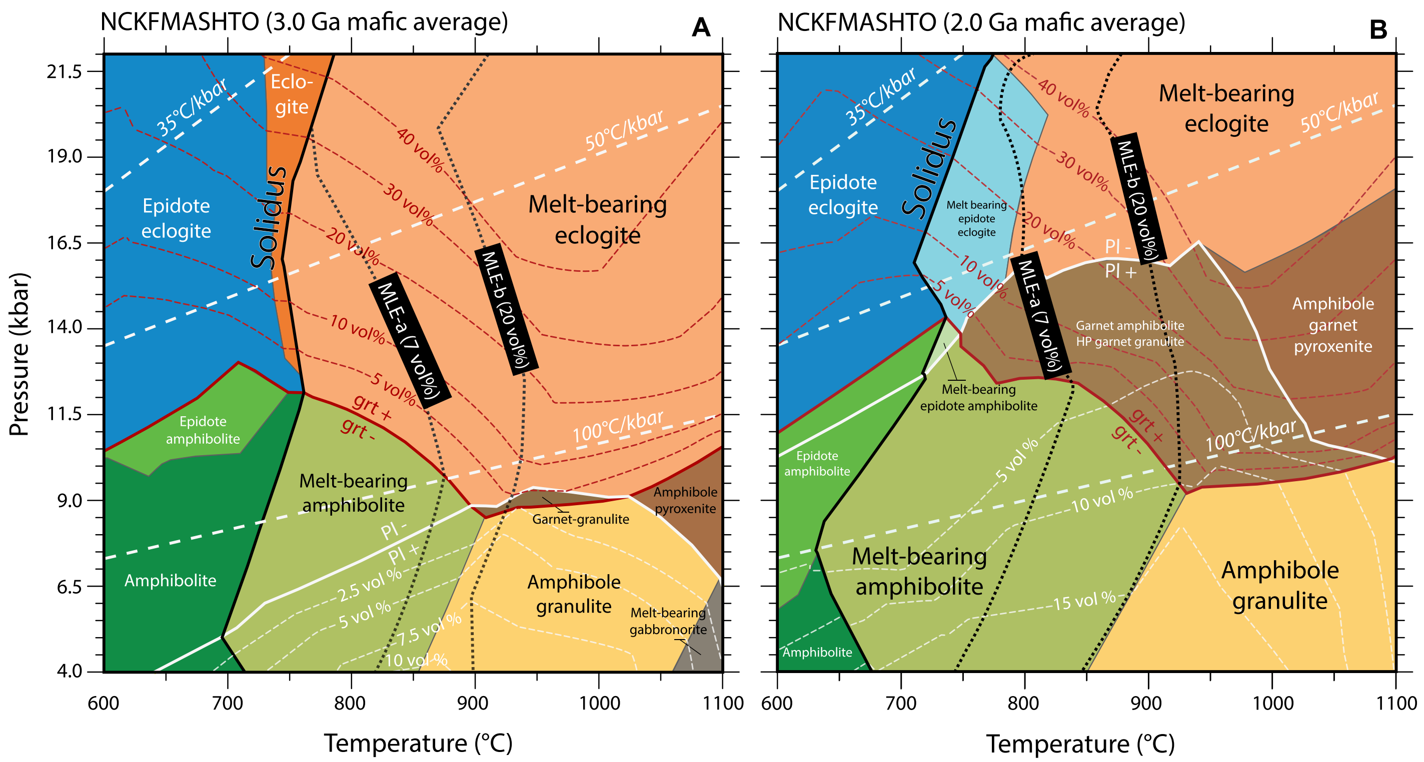


**Figure 2. (A,B) Calculated pressure-temperature (*P-T*) phase diagrams (lithological) for mafic crust composition at 3.0 Ga (A) and 2.0 Ga (B) in the NCKFMASHTO ($Na_2O$–CaO–$K_2O$–FeO–MgO–$Al_2O_3$–$SiO_2$–$H_2O$–$TiO_2$–$Fe_2O_3$) chemical system (see Supplemental Material S2 [see footnote 1] for details on thermodynamic modeling). Diagrams show effects of cogenetic phases on melt composition. Two melt loss events (MLEs) are shown at 7 vol% and 20 vol% melt (black dashed lines). Geotherms are displayed as bold dashed white lines, and phase proportions in vol% are shown as light dashed lines in red for garnet (Grt) and in white for plagioclase (Pl). HP—high pressure.**

the plagioclase-out line at 16 kbar at 950 °C (Fig. 2B). This suggests that more than 9 vol% of plagioclase would be produced as a cogenetic phase at MLE-a and MLE-b conditions under the high d*T*/d*P* gradient. No plagioclase would be stable under the intermediate d*T*/d*P* geothermal gradient.

Based on thermodynamic modeling, we suggest that the higher negative Eu anomalies observed in the 2.0 Ga zircon population are a function of changes in the average mafic crust composition and, hence, the composition of the derived intermediate to felsic melt in which zircons grew in equilibrium (Fig. 3). As Na, Eu, and Sr are all preferentially partitioned into plagioclase, this observation would also have implications for the evolution of the whole-rock Rb/Sr ratio and the global differentiation of the crust (Dhuime et al., 2012), without invoking major variations in the crustal geotherms or in the general crustal thickness and the depth of magmagenetic sites. Alternatively, Stern (2020) proposed that melts produced during a single-lid Mesoproterozoic tectonic regime (1600–1000 Ma) would be less hydrous (i.e., lower oxygen-fugacity conditions), which could also contribute to low Eu/Eu* values in Proterozoic zircons, although the drop in Eu anomaly occurred prior to this period in Earth history.

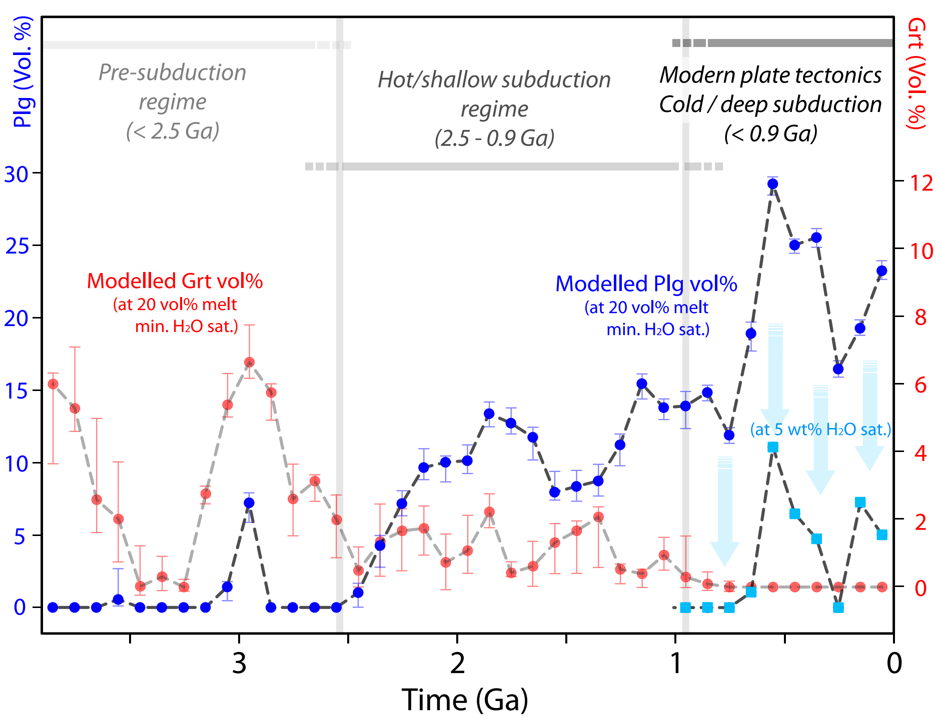


**Figure 3. Time series of modeled volumetric proportions of garnet (Grt, in red) and plagioclase (Plg, blue dots) phases at melt loss event B (20 vol% of melt produced along 100 °C/kbar geotherm) under minimum water-saturation conditions and at 5 wt% $H_2O$ saturation (Plg, light blue squares), using successive average mafic composition for each 100 m.y. step (in the NCKFMASHTO [$Na_2O$–CaO–$K_2O$–FeO–MgO–$Al_2O_3$–$SiO_2$–$H_2O$–$TiO_2$–$Fe_2O_3$] chemical system; see details in Supplemental Material S2 [see footnote 1]).**

**Are Post–900 Ma Magmagenetic Sites Becoming Superhydrous?**

Starting at the beginning of the Neoproterozoic, the global zircon Eu anomaly became less negative, continuing this trend from 900 Ma to present day (Fig. 1). This trend is marked by the development of lower d*T*/d*P* metamorphic thermal gradients around 900–800 Ma (30–35 °C/kbar) and the general development of "modern" subduction systems (also referred to as "cold subduction zones"; Brown and Johnson, 2018) and the widespread occurrence of modern plate tectonics (Cawood et al., 2018). The Mesoproterozoic-Neoproterozoic transition coincides with a decreasing Rb/Sr ratio in juvenile crustal rocks, interpreted as a decrease in average crustal thickness (~30 km) and the point at which crustal destruction rates may have exceeded the rates of juvenile crust production (Dhuime et al., 2012). A decrease in global crustal thickness from the Neoproterozoic to present day is also supported by the evolution of pyroxene and amphibole compositions and Ca contents in mafic lava (Ganne et al., 2018).

Using the 1.0 Ga average mafic composition, the *P-T* phase diagram (Fig. S1) shows that the garnet-in line is equivalent to that in the 2.0 Ga phase diagram (i.e., garnet-in at 9 kbar around 900 °C; see Fig. S1). For the diagram corresponding to the 0.05 Ga composition, the plagioclase stability field extends up to 20 kbar at 1050 °C, whereas the garnet-in reaction line shifts to higher *P-T* conditions (14 kbar at 900 °C; Fig. S2). This suggests that the less negative Eu anomaly in zircon after 900 Ma could be explained by the general development of a lower geothermal gradient. A lower geothermal gradient suggests that magma generation would take place at higher pressure with a lower modal proportion of plagioclase and, therefore, in a thicker crust environment.

The global development of "colder" subduction systems subsequent to 1.0 Ga suggests an increase of water and other volatiles in the mantle that could trigger hydrous melting (Stern, 2018), favoring water saturation and more oxidizing conditions at deep crustal levels. However, the effect of pressure and water content on zircon saturation in felsic and intermediate melts is relatively minor (e.g., Boehnke et al., 2013). In contrast, plagioclase saturation at a given pressure can be delayed to lower temperature in hydrous mafic systems or even suppressed for superhydrous conditions at middle- to lower-crustal pressures (>3 wt% $H_2O$; Müntener et al., 2001).

We modeled the impact of water on the average mafic rock composition at 1.0 Ga under suprasolidus conditions (at a constant *T* of 900 °C, at which melt is present; see Figs. S1 and S2). Our modeling showed that the plagioclase mode drastically decreases with increasing water content, with the plagioclase-out line at 12 kbar for 3.2 wt% $H_2O$ (Fig. 3). The water content of mafic to intermediate magmas from active magmatic arcs is, on average, 4.0 ± 1.3 wt% and reaches up to 7–8 wt% based on recent compilations (Rasmussen et al., 2022). We thus suggest that the less negative zircon Eu anomaly could also mark a general increase in water content—and oxidizing conditions—at magma genesis sites since 900 Ma, which may coincide with the onset of modern cold subduction zone systems and modern plate tectonics.

## CONCLUSIONS

How and when plate tectonics began on Earth and Earth's tectonic style before this geodynamic development remain outstanding geoscientific questions. Due to its resilience and ubiquity in the geologic record, zircon trace-element geochemistry provides a robust archive of magmatic and tectonic processes. We present a statistical analysis of the zircon Eu anomaly record from 3.2 Ga to present in order to resolve global geodynamic processes. Previously, the Eu anomaly in zircon has found application as a potential proxy for crustal thickness. However, crustal thickness is only one of many (underestimated) factors that may influence zircon geochemistry, e.g., magmatic source composition, redox and *P-T* conditions, water content. We identified two major changes in the global record of zircon Eu anomalies, a ca. 2.5 Ga decrease and a ca. 0.9 Ga increase. We attribute the 2.5 Ga decrease to secular evolution in the compositions of the source magmas from which these zircons were derived, whereas we associate the 0.9 Ga increase in the zircon Eu anomaly to the global development of cold subduction systems and the onset of modern plate tectonics.

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