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Europium traces the impact of high temperature hydrothermal systems on the early oceans

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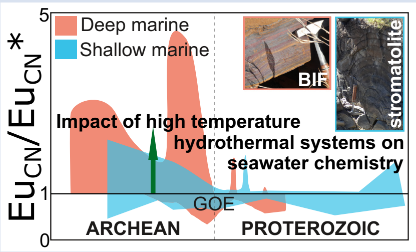
Abstract



Hydrothermal systems have been invoked as a major driver for the evolution of life, but the impact of hydrothermal fluids on Earth's ancient oceans and their habitats remains ambiguous. Europium (Eu) enrichments trace high temperature hydrothermal fluids in rock archives and may serve as proxy for hydrothermal input into ancient oceans. Here, we provide Eu abundances from stromatolites and iron formations between 3.8 and 0.542 billion years (Ga) ago and reconstruct the impact of hydrothermal systems on shallow and deeper marine environments. Our results document a continuous decrease in positive Eu anomalies until 2.5 Ga ago, followed by almost complete disappearance, suggesting a decreasing impact of submarine hydrothermal systems on ancient oceans. Exceptional positive Eu excursions between 2.8 and 2.6 Ga, and potentially also at 3.5 and 2.2 Ga, are only preserved in deep marine settings and reflect magmatic pulses triggered by elevated upper mantle temperatures. Our results demonstrate the significance of high temperature hydrothermal systems on Archean seawater chemistry with implications for the supply of bio-essential elements. However, life in shallow marine environments was likely supported by fluxes from emerging continents, at the least from the Neoarchean onwards.

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Introduction

Environmental conditions during the origin and early evolution of life on Earth are intensely debated. Hydrothermal systems have long been considered critical players in this debate because they may have released essential nutrients and provided catalytic minerals for the formation of prebiotic organic molecules (*e.g.*, Martin *et al.*, 2008). Hydrothermal settings have been described from the early Archean rock record (Vearncombe *et al.*, 1995; Djokic *et al.*, 2017) and are implicated in models for the precipitation of iron formations 3.8–1.8 Ga ago (Isley and Abbott, 1999; Tosca and Tutolo, 2023). However, a quantitative assessment of the intensity and fluctuations of mass fluxes originating from hydrothermal systems contributing to the elemental inventories of early oceans has so far not been attempted.

Europium can fill this gap. It occurs in a 3+ state under surface environmental conditions similar to the other rare earth elements (REE) but is reduced to the more soluble Eu^{2+} under reducing, acidic, conditions above 250 °C, and enriched relative to the neighbouring REE (Bau, 1991). Thus, high temperature hydrothermal fluids become enriched in Eu relative to the host rocks during fluid–rock interactions. Although Eu^{2+} immediately oxidises to Eu^{3+} in lower temperature regimes, where it behaves geochemically similar to the neighbouring, strictly trivalent REE, the precipitates of hydrothermal fluids can preserve this Eu enrichment over geologic time. In the modern fully vented and oxidised oceans, REE are particle-reactive and show residence times of only a few hundred years. Acidic fluids reaching temperatures up to ~400 °C with positive Eu anomalies are exhaled at hydrothermal vent systems such as submarine trenches, hot spots, or rift systems into seawater, and are immediately cooled and oxidised (*e.g.*, German *et al.*, 1990). Subsequently, precipitating Fe- and Mn(oxy)hydroxides scavenge REE from the hydrothermal fluid and seawater. Therefore, hydrothermal Fe-Mn oxide precipitates (Bau *et al.*, 2014) and carbonates in mussel shells proximal to black smoker vent sites show strong Eu enrichments (Bau *et al.*, 2010). Modern submarine high temperature hydrothermal systems, where Fe-Mn oxides precipitate proximal to the vent site, act as sinks rather than sources for REE and other particle-reactive elements (German *et al.*, 1990). In a stratified, anoxic water column, such as proposed for long periods of the Precambrian, REE could

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likely reach much longer residence times due to the lack of direct scavenging onto Fe- and Mn(oxy)hydroxides. Thus, REE exhaled from hydrothermal systems into Precambrian oceans should have been dispersed more widely. However, beyond computational models (Tutolo and Tosca 2023), there is no empirical constraint on the extent to which these fluids contributed to water chemistry in the different environments of early oceans, such as outer shelf or near shore environments.

Europium systematics have previously been used to investigate the impact of high temperature, hydrothermal systems on ancient local depositional environments (*e.g.*, Bau and Dulski, 1996) and the thermal state of Earth's mantle (Danielson *et al.*, 1992; Viehmann *et al.*, 2015), as well as in ore geology to fingerprint the temperature of ore-forming hydrothermal fluids (Kraemer *et al.*, 2019). Prior studies of iron formations (IFs; Danielson *et al.*, 1992; Viehmann *et al.*, 2015) have shown a decline of Eu anomalies between 3.8 Ga and 2.7 Ga ago, with a much more positive Eu peak at 2.6 Ga. These studies proposed variable REE fluxes from submarine high temperature systems into Archean seawater.

While these studies demonstrate that Eu signatures in chemical sediments can serve as a robust proxy for hydrothermal inputs, the previous focus on IFs limits current knowledge to the deeper continental shelf and, in some cases, to sites that were deposited in proximity to active submarine volcanic zones. A new approach is needed to track hydrothermal influences on shallow marine settings, which are known for occurrences of stromatolites from *ca*. ≥3.5 Ga until today (Riding, 2011) and have been crucial for the appearance and radiation of oxygenic phototrophs that ultimately led to the oxygenation of Earth's atmosphere at *ca*. 2.4 Ga (Lyons *et al.*, 2014). Shallow marine sites also host the earliest eukaryotic algae in the Proterozoic (Javaux *et al.*, 2001). Accurate reconstructions of the evolution of Earth's biosphere and surface environments thus require an assessment to establish if, and to which extent, hydrothermal systems in these shallow marine settings affected water chemistry and the delivery of nutrients amongst other elements.

Here, we report Eu anomaly curves of IFs and marine stromatolites throughout the Precambrian to determine the influence of high temperature hydrothermal fluxes on seawater chemistry in both deep and shallow marine environments. The results allow constraint of the impact of high temperature hydrothermal systems on a range of marine habitats throughout Earth's history.

The Link of Eu Abundances in Iron Formations to the Geodynamic Evolution of the Earth

The compilation of chondrite-normalised Eu mass fractions (Eu_{CN}) in IFs (Fig. 1) and stromatolites (Fig. 2) reflects deep and shallow marine Precambrian environments. The enrichment of Eu in carbonates, where REE substitutes for Ca in the crystal lattice, is not quantitatively comparable to REE enrichments in IFs that are the result of sorption onto Fe(oxy)hydroxides. However, qualitative trends of Eu anomalies in both rock types reflect the change in concentration of Eu in the Precambrian oceans over time. Our data compilation includes new and published data, screened for sample purity. Notably, all samples used for Eu anomaly evaluation show shale-normalised, seawater-like REE patterns with positive La, Gd, and Y anomalies, as well as a heavy over light REE enrichment. This rules out significant amounts of detrital contamination and post-depositional REY mobility during diagenesis, metamorphism, or weathering that would instead result in non-seawater-like REE signatures (see Methods in the Supplementary Information). Sample details are available in the Supplementary Information.

Our new compilation of IFs shows consistently positive Eu abundances throughout the Archean, irrespective of IF type (Algoma-type IFs are related to volcaniclastic sequences, whereas Superior-type IFs were deposited on the continental shelf and slope; *e.g.*, Gross, (1965). Within the Archean, the Eu curve shows an overall decrease from the Eoarchean until *ca*. 2.8 Ga, followed by a strong positive Eu excess between 2.8 and 2.55 Ga that is larger than in the Eoarchean (Fig. 1). High Eu anomalies in the Eoarchean are related to a hotter upper mantle resulting from the combination of residual accretionary

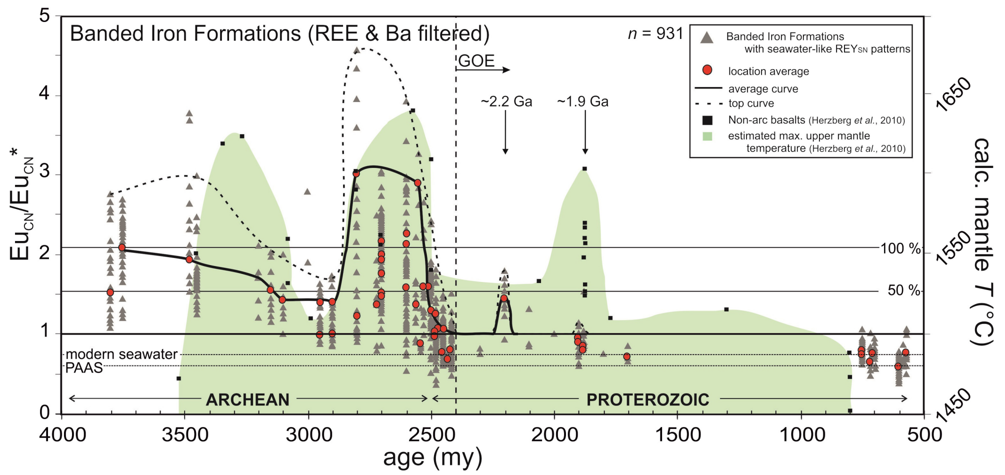

Figure 1 Precambrian Eu_{CN}/Eu_{CN^*} evolution curve of iron formations. The dashed and solid lines represent the global top and location average Eu_{CN} anomaly curves of pure iron formation samples (931 data points). Eu_{CN} anomalies are abundant throughout the Archean and decrease from the Eoarchean until *ca*. 2.5 Ga ago. Positive Eu_{CN} excursions from 2.8 until 2.55 Ga and potentially at ~2.2 and ~1.9 Ga suggest strong REE fluxes into the oceans due to intense magmatic activity. Note that the Eu_{CN} evolution curves coincide with the evolution of calculated upper mantle temperature values (black squares and green field with estimated mantle temperatures on the right y-axis; Herzberg *et al*., 2010). Percentage values refer to the de- or increase of Eu_{CN}/Eu_{CN^*} values relative to the Eoarchean Isua BIF; the location average includes >3 samples. Respective data is available in the Supplementary Information. GOE: Great Oxidation Event; PAAS: Post Archean Australian Shale.

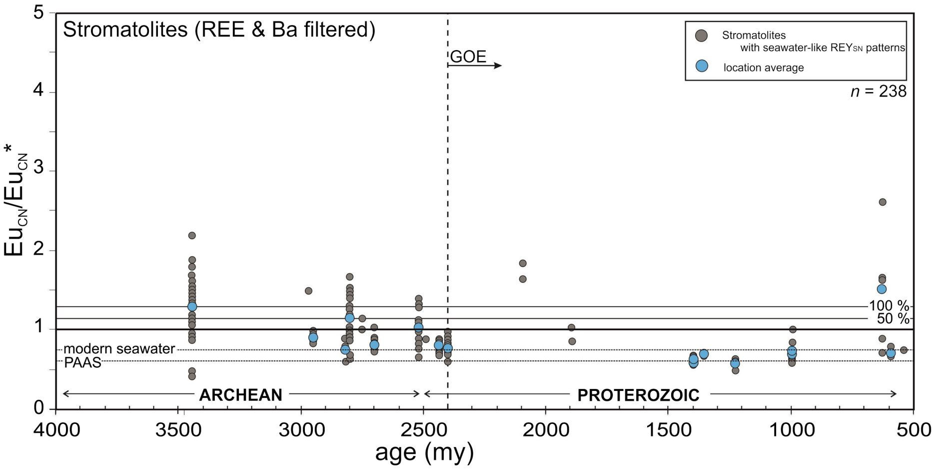

Figure 2 Precambrian Eu_{CN}/Eu_{CN*} evolution curve of stromatolites deposited in marine environments. The average value of Eu_{CN} anomalies in pure stromatolites (229 data points), representing shallow marine microbial habitats, decreases from the Palaeoarchean until 2.5 Ga. Eu_{CN} anomalies are generally absent in the Proterozoic with two local exceptions at 2.1 and 0.635 Ga. Note that some of the Archean stromatolite locations do not show positive Eu_{CN} anomalies. Percentage values are relative to Palaeoarchean stromatolites from the Pilbara Craton. Location average includes >3 samples. Respective data is available in the Supplementary Information. GOE: Great Oxidation Event; PAAS: Post Archean Australian Shale.

heat and a higher abundance of radioactive elements (Korenaga, 2008). At *ca.* 3.8 Ga, average Eu anomalies in the Isua supracrustal belt (Greenland) are more positive than those observed in Nuvvuagittuq greenstone belt (Canada), maybe due to variable upper mantle temperatures that led to local Eu enrichments and depletions in seawater at that time. The decrease in Eu anomalies from the Eoarchean until 2.8 Ga (~80 %) is interpreted to reflect overall decreasing upper mantle temperatures and, therefore, a decreasing impact of high temperature hydrothermal systems on ocean chemistry.

A strong Eu_{CN}/Eu_{CN}* peak, on average *ca.* 20 % higher than initial Eoarchean values, occurs in multiple localities between 2.8 and 2.55 Ga. The starting point of this period is locally preserved in the *ca.* hundred-million-year older Kushtagi Shist Belt of the Dharwar Craton with up to 87 % higher Eu_{CN}/Eu_{CN}* values than in the Eoarchean; the termination of this period is recorded in the 2.55 Ga old Nanfen IF with up to 76 % higher Eu_{CN}/Eu_{CN}* values. Herzberg *et al.* (2010) estimated upper mantle temperatures of *ca.* 1500 to 1600 °C between 3.0 and 2.5 Ga derived from non-arc basalts. Interestingly, the Eu_{CN} anomalies of Archean seawater mirror the estimated upper mantle temperature evolution (Herzberg *et al.*, 2010; Fig. 1). Their data show a progressive decrease from >1600 °C to <1550 °C between 3.4 and 2.8 Ga, followed by a return to almost 1650 °C between 2.8 and 2.5 Ga (Fig. 1). If this comparison is valid, Eu_{CN} systematics in IFs are a promising proxy to estimate upper mantle temperatures. Conversely, our data identify a strong linkage between solid Earth processes and global ocean chemistry.

Positive Eu_{CN} anomalies in IFs nearly disappear from the rock record by the onset of atmospheric oxidation at *ca.* 2.4 Ga, known as the Great Oxidation Event (GOE). From this point onwards, Eu_{CN}/Eu_{CN}* values are below unity and cluster around the value of modern seawater, suggesting insignificant impact of high temperature hydrothermal systems on the deep marine chemistry of REE and other particle-reactive elements. There are two possible reasons for this drop. First, mantle temperatures and resultant hydrothermal activity may have declined asymptotically (*e.g.*, Labrosse and Jaupart, 2007), so that Eu signals from hydrothermal vents became too dilute in the global ocean to leave a trace in IF. Second, travel distances of water masses carrying a hydrothermal signature declined due to the penetration of O_2 into the water column (Poulton and Canfield, 2011) that led to the formation of Fe- and Mn(oxy)hydroxides and scavenging of REE deeper in the oceans and a lower residence time of REE. Both scenarios, or a combination thereof, would have led to an almost complete dilution of Eu_{CN} anomalies in Proterozoic IFs. Periodic, local peaks in Eu_{CN} anomalies at 2.2 Ga and around 1.9 Ga indicate a temporary return to intense magmatic activity and higher upper mantle temperature that led to elevated REE fluxes into seawater via high temperature hydrothermal systems. Notably, these Proterozoic Eu_{CN} excess events and the broad Eu_{CN} anomaly peak between 2.8 and 2.6 Ga overlap with proposed mantle overturn events tied to supercontinent aggregation (Bédard, 2018), which are also thought to have triggered widespread IF deposition (Isley and Abbott, 1999). Other Proterozoic thermal peaks around 1.0 and 0.6 Ga ago (*cf.* Arndt, 2013) are not recorded in IFs, probably related to the above-mentioned scenarios leading to lower REE residence times and/or IF preservation bias due to their scarcity in the Proterozoic record. Importantly, Neoproterozoic IFs show no evidence for contributions from high temperature hydrothermal systems due to widespread and deeper ocean ventilation (Baldwin *et al.*, 2012), leading to rapid REE scavenging and shorter residence times of REE in the global ocean. By this point, residence times of REE were perhaps within a few hundred years, comparable to those of modern oceans.

Impact of High Temperature Hydrothermal Fluids on Shallow Marine Precambrian Microbial Habitats

Marine stromatolitic carbonates are mostly preserved as lime- or dolostones in worldwide occurring Precambrian stratigraphies and provide, independent of their mineralogy, a local snapshot into near shore microbial habitats within the photic zone. Eu_{CN} anomalies in such archives suggest that high temperature hydrothermal systems impacted these environments, which may potentially have influenced the evolution of phototrophic organisms commencing at ~3.5 Ga (Fournier *et al.*, 2021). The largest Eu_{CN} anomalies are preserved in the oldest stromatolitic carbonates from the Pilbara Craton, suggesting the strongest impact of hydrothermal influx on local seawater chemistry in these habitats (Fig. 2; Van Kranendonk *et al.*, 2019).

Positive Eu_{CN} anomalies are also observed in some Meso- and Neoarchean stromatolites, reflecting locally or temporarily enhanced hydrothermal input to these sites. However, many Archean stromatolite locations lack positive Eu_{CN} anomalies, indicating that hydrothermally-derived REE from high temperature systems, as captured by IF, were efficiently scavenged in deeper marine settings or diluted by continentally derived solutes. Hence, hydrothermal REE fluxes may not always have reached near shore, shallow marine microbial habitats during the Archean. Overall, maximum Eu_{CN} anomalies decrease over time across the Archean, but there is no distinct enrichment between 2.8 and 2.55 Ga as recorded in the IF data, suggesting that hydrothermal fluids during this intense magmatic period had no impact on the REE budget of Neoarchean near shore, shallow marine environments.

Interestingly, the unfiltered stromatolite Eu_{CN} anomaly curve (Fig. S-3 in the Supplementary Information) shows positive peaks between 2.9 and 2.7 Ga, 2.1 Ga, and 1.9 Ga. Most of these positive Eu anomalies are artificial based on BaO interference on Eu concentration during analyses (see Supplementary Information); the high concentrations of Ba in the carbonates, however, may hint at temporal increased Ba-rich fluxes from hydrothermal systems into seawater. Although Eu_{CN} anomalies are present in some Archean stromatolites, the impact of high temperature hydrothermal systems on seawater chemistry in stromatolite-forming microbial habitats was only locally elevated. Similarly, positive Eu_{CN} anomalies in the Neoproterozoic are almost entirely absent in the IF record except for a few stromatolites from the Taoudeni Basin. Thus, microbial life thriving in habitats without influx from hydrothermal systems had to adapt to element delivery from different sources, such as continental solutes. This is supported by the complete absence of Eu_{CN} anomalies in half of the Archean stromatolite localities, providing evidence that microbial life in photic zones of near shore environments was probably not dependent on element fluxes of REE and other elements with comparable residence times from high temperature systems, but instead was reliant on solutions derived from weathering and erosion processes of already emerged landmasses.

Significance of Bio-Essential Elements Delivered From High Temperature Systems Into Archean Oceans

Armed with a new constraint on the influence of high temperature hydrothermal vents on seawater chemistry through the Precambrian, we independently explored in a first order model the implications of hydrothermal activity for bio-essential elements, such as Mn, Co, Ni, Cu, and Zn that are enriched in hydrothermal fluids compared to seawater and river waters (Von Damm, 1995). To constrain the hydrothermal flux of each of these elements, we mixed a black smoker fluid with modern and Archean seawater, respectively, using a thermodynamic model (see Supplementary Information for details). The final amount of the respective element in solution, after precipitation of hydrothermal precipitates, was multiplied by the hydrothermal water flux. To be conservative, we assumed that the hydrothermal flux was the same as today, but as shown by the Eu data, the flux may have been much higher during parts of the Archean. The riverine element flux was calculated from the river water flux multiplied by average elemental concentrations in modern and Archean river water. The results suggest that, even without additional enhancement above modern levels, hydrothermal vents

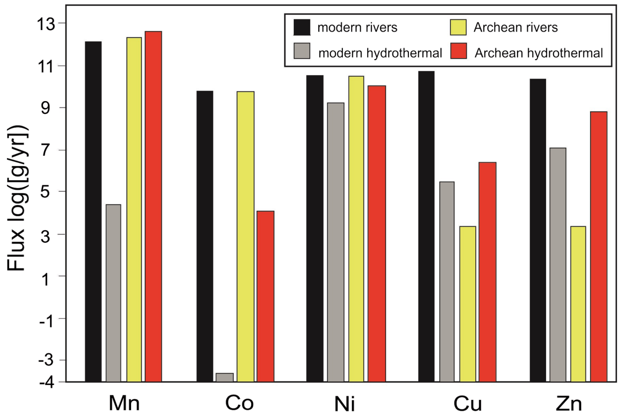

Figure 3 Flux estimate of bio-essential elements via rivers and hydrothermal systems. See Supplementary Information for model assumptions and parameters.

would have been the dominant sources of Mn, Cu, and Zn to the Archean ocean (Fig. 3). If the river water flux was smaller due to smaller landmasses while hydrothermal water fluxes were elevated, as suggested by the Eu_{CN} data for extended times during the Archean (Fig. 1), also the input of Ni would likely have been dominated by hydrothermal sources.

Today, rivers constitute the major sources of all of these dissolved elements to the ocean (Fig. 3). The first evidence of continental weathering affecting seawater chemistry is reported at *ca.* 3.2 Ga ago (Satkoski *et al.*, 2016), and it became widespread in the Neoarchean at around 2.7–2.6 Ga ago (Viehmann *et al.*, 2014, 2018). We, therefore, speculate that the transition to riverine dominance in nutrient fluxes dates back to the early Proterozoic, when continental land masses were likely well established while the hydrothermal influence waned (Fig. 1). Life in the Neoarchean, in the lead-up to the Great Oxidation Event, may have benefitted from a combination of elevated hydrothermal activity (Fig. 1) and increasing continental weathering (Viehmann *et al.*, 2014, 2018). In conclusion, our data suggest that the fluxes of hydrothermally sourced bio-essential elements into the Archean oceans were most potent in the Eoarchean, around the time of life's origins, and again in the Neoarchean when nutrient fluxes drove cyanobacterial diversification and niche expansion (Fournier *et al.*, 2021). It is, therefore, conceivable that elevated mantle temperatures and resulting hydrothermal fluid flow indirectly drove biological activity and innovation by modulating nutrient supplies.

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Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2514.

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