## Secular change in the Si isotope compositions of TTGs indicates increased weathering of subaerial land ~3.7 billion years ago

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Multiple lines of evidence suggest that 25-75% of continental crust already existed by 3.0 Ga<sup>1-2</sup> but it is not clear how much of this crust sat above sea-level; nor is it clear how early in Earth's history the realm of subaerial continental crust became established. Constraining this is important, because weathering and erosion of emerged lands plays a significant role in the Earth system, exerting a control on the composition of the atmosphere and nutrients delivered to the oceans. Furthermore, emerged land hosts lakes and continental shelves that are thought to have provided ideal habitats for the emergence of life<sup>3</sup>.

Studies have shown that the silicon isotope compositions of Archaean granitoids are resolvably heavier than their Phanerozoic equivalents, due to the presence of marine authigenic silica in the source region of TTG melts<sup>4-5</sup>. This silicified material provides a proxy for the Si isotope composition of Earth's early oceans and thus records the balance of sources and sinks. We show that the Si isotope compositions of TTGs changed geologically rapidly around 3.6 Ga - and suggest that the most likely explanation for this is an increase in isotopically heavy dissolved silicon derived from subaerial weathering of continental crust. Modelling of this shift reveals that between 3.8 and 3.6 Ga, the relative magnitude of terrigenous input to the marine dissolved Si budget increased from near zero to  $32 \pm 15$  %. This would suggest that, from 3.6 Ga onwards, continental weathering feedbacks were established and mass flux from land became an important source in the chemical budget of seawater - changes that likely exerted a positive effect on the evolution of life.

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