

The Neoproterozoic surface sulphur cycle: New solutions to non-zero intercept $\Delta^{33}\text{S} - \delta^{34}\text{S}$ arrays

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We report *in situ* SIMS S-isotope data for sedimentary pyrite from the 2.52 Ga Upper Campbellrand Subgroup, Transvaal, South Africa. The analysed rock is a shaly limestone from the Gamohaana Formation interpreted to have been deposited in a water depth of ca. 50-100 m, in a restricted basin. A previous study documented that in $\Delta^{33}\text{S}$ vs. $\delta^{34}\text{S}$ space, the pyrites define a strong linear trend with a significant offset from a zero intercept. The present study revisited the sample, carrying out further analyses of pyrite. The new data confirm and expand this non-zero intercept, linear array intersecting ca. $\delta^{34}\text{S}$ of 0 at $\Delta^{33}\text{S}$ of +5. One interpretation of this array is that it indicates mixing of unrelated S-sources in the sedimentary environment, specifically recycled S from sulphides of sulphate reducing bacteria origin, along with elemental S. This mixing generated isotope compositions of sulphides plotting to the left of the atmospheric array at a $\Delta^{33}\text{S}$ value intermediate between the end-member H_2SO_4 and S_8 components.

Alternatively, the non-zero intercept array may provide evidence that, at least temporarily, the sampled atmospheric S had a different composition from bulk Earth, indicating a very localised S-isotope signature in the Neo-archaeon surface environment. This alternative hypothesis is supported by two observations. Firstly, from an analogy with Pb whose low residence time and low concentration in the modern ocean-atmosphere system, analogous to S in the Archaeon, permit effective transfer and preservation of the localised industrial atmospheric pollution signal into sediment. Secondly, because the predominant Archaeon source of seawater sulphate was volcanogenic, the evolving nature of terrestrial magmatism, particularly the onset of subduction, must have influenced the quantity and species of S eventually incorporated into sediments. Thus, the hypothesis tested in this study is whether the large late Archaeon 'global' S-MIF excursions and asymmetry can be explained by superposition of extreme local signals. Fallout from local volcanic eruptions could imprint MIF-S signals into pyrite of restricted basins, thereby avoiding blurring the signal in the subdued, averaged global open ocean S pool, in the lead-up to a new tectonic framework that culminated in the Archaeon-Proterozoic boundary.