Organic-bound trace metals and geosphere-biosphere co-evolution: Development of a novel biosignature

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The study of C-N-S isotopes is a well-established approach for investigating the interplay between environmental changes and the Earth's early biosphere. Here we introduce the development of a novel biosignature, by extending the traditional geochemical approach to the abundance and isotopic systems of organic-bound trace metals.

Metalloproteome analyses suggest that trace metals (e.g. Cu, Fe, Mo, Ni, Zn) were selectively utilised by different organisms and metabolisms at different times throughout the evolution of the biosphere. We seek to investigate this hypothesis by extracting trace metals from the organic matter (kerogen) preserved in a suite of rocks and sediments spanning the past 3.5 billion years. We have developed an extraction methodology that enables us to successfully determine and compare the abundances and isotopic signatures of trace metals within the silicate fraction (or bulk rock), the sulphide fraction, and the purified kerogen fraction of each individual sample. This has allowed us to observe distinct differences in trace metal geochemistry both between fractions of individual samples and between the same fractions from samples of different ages.

Our preliminary results from the Archean Kaapvaal Craton, South Africa and modern Peru Margin sediments are especially striking in the case of Fe and Zn and their isotopes. Our data agrees with previous work [1] in highlighting light Fe in the Archean versus heavier Fe in modern sulphides. An opposite trend is observed for Zn with respect to Fe. Our data suggest fundamentally different patterns of oceanic mass balance and biospheric uptake for the Archean versus the modern, and shows promise for elucidating the currently unresolved Zn oceanic mass balance [2]. Ultimately, we show that the application of trace metal geochemistry has great potential as a novel biosignature, likely to be of instrumental importance in understanding the co-evolution of the Earth's dynamic geosphere and biosphere.

[1] Anbar A.D. and Rouxel O.J. (2007) *Annu. Rev. Earth Planet. Sci.* **35**, 717-746. [2]Little S.H., et al. (2014) *GCA* **125**, 673-693.