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Reconstructing global and local sulfur cycle evolution using Carbonate Associate Sulfate in ancient sedimentary strata

GUILLAUME PARIS[1], ARBIA JOUINI[2], VICTORIA C.F. BLACK[3], ALEXANDRA (SASHA) V. TURCHYN[4], DAN JOHNSON[5], JESS ADKINS[6] AND THEODORE PRESENT[6]

[1]CRPG (Université de Lorraine - CNRS)
[2]CRPG-CNRS
[3]Not affiliated
[4]University of Cambridge
[5]Rice University
[6]California Institute of Technology

Presenting Author: guillaume.paris@univ-lorraine.fr

The isotopic composition of sulfur in CAS (Carbonate-associated sulfate) is frequently used to reconstruct the isotopic composition of dissolved sulfate in ancient oceans, which itself reflects changes in the biogeochemical sulfur cycle. One of the most striking feature of the sulfur isotopic composition of CAS is a drastic decrease in isotopic range and variability over the Phanerozoic, often interpreted as reflecting an increase in seawater sulfate concentration. What is less often considered is the impact of sedimentary diagenesis on the measured sulfur isotopic composition of CAS.

In sediments, microbial sulfate reduction modifies the isotopic composition of sulfate. If the isotopically modified sulfate is incorporated into carbonate minerals, the measured sulfur isotopic composition of CAS would reflect not that of seawater but instead that acquired in the sediment during diagenesis. Over the last ten years, theoretical and analytical progress has helped us acquire a stronger understanding of how CAS works as a proxy. Overall, previous results showed that isotopic heterogeneity exists within the carbonate components of a rock. We also demonstrated that CAS in well-preserved biogenic foraminifera calcite can provide records as reliable as pelagic barite over the last 60 My. However, when working in older strata, such comparison is not necessarily possible.

Here we present published and new records of variations in the sulfur isotopic composition of CAS at the hand-sample scale, and demonstrate that all carbonate phases within a sample do not react identically and that diagenesis can affect CAS in a more complicated way than well-studied mechanisms of burial and meteoric diagenesis. This realization opens the way both to an increased reliability of the carbonate record for reconstructing past seawater sulfur isotopic composition and to extracting new information from the sedimentary record. Thanks to highly precise small scale sulfur isotopic analyses, the reconstruction of microbial processes within ancient sediments might be the way to further understand past biogeochemical cycles, unlocking new constraints on the carbon and oxygen cycles through time, and eventually unravelling the reasons behind the decrease in CAS sulfur isotope variability through the Phanerozoic.