Goldschmidt 2026 Abstract
https://doi.org/10.46427/gold2026.37162

New Insights into Sulfur Cycle–Sulfur Isotope Geochemistry: The Geochemical Behavior of Volatile Organosulfur Compounds (VOSC)

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In the modern ocean, sulfate has an exceptionally long residence time of approximately 13 million years, with a highly uniform sulfur isotope composition in non-sulfidic seawater. This uniformity is the cornerstone for using sulfur isotope proxies to reconstruct seawater sulfur isotope composition and constrain the sulfur cycle. However, in waters with low sulfate concentrations and widespread sulfidic conditions, inorganically derived volatile organosulfur compounds (VOSC) can significantly alter the sulfur isotope composition of non-sulfidic seawater. Previous studies have used this mechanism to explain the abundant superheavy pyrite formation in the Neoproterozoic.

We compiled the evaporite sulfur isotope record and the carbonate-associated sulfur (CAS) record since 600 Ma. Although both are used to reconstruct seawater sulfur isotope composition, they exhibit systematic differences. This difference (ΔEva-CAS) shows a decreasing trend over time: from positive values (evaporite sulfur isotope values higher than CAS) before the Paleozoic Oxidation Event (POE) , to statistically insignificant differences during the POE, to evaporite values lower than CAS afterward.

Through a one-dimensional diffusion-advection-reaction model, we revealed a key point overlooked by previous studies. Because seawater advection and turbulent diffusion equally affect all dissolved components, the total dissolved sulfur content and sulfur isotope composition in seawater remain conserved—regardless of sulfate and hydrogen sulfide interconversion—as long as non-dissolved forms (e.g., gaseous and solid phases) are not produced. Therefore, the downward-lightening sulfur isotope gradient in non-sulfidic seawater indicated by positive ΔEva-CAS values cannot result from early diagenetic reactions in carbonates or hydrogen sulfide oxidation, but instead indicates an active VOSC cycle.

The model and static results further show that this gradient corresponds to superheavy pyrite deposition. Positive ΔEva-CAS values indicate VOSC production and its oxidation in non-sulfidic seawater, with the active VOSC cycle being the core mechanism for abundant superheavy pyrite deposition.

Before the POE, a Dissolved Organic Carbon (DOC) reservoir in seawater enabled inorganically derived VOSC production in sulfidic conditions. During and after the POE, complete oxidation of deep seawater, reduction of the DOC reservoir, and disappearance of sulfidic seawater rendered the inorganic VOSC cycle inactive, establishing the sulfur cycle and isotope recording pattern of the modern ocean.

