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Decoding Earth's Redox History: High-Dimensional Isotope Systematics in Carbonate-Associated Sulfate

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Carbonate-associated sulfate (CAS)—trace sulfate ions structurally incorporated into the crystal lattice of carbonate minerals—serves as a premier archive of ancient seawater sulfate chemistry, directly linking the biogeochemical cycles of sulfur, oxygen, and carbon. The recent emergence of high-dimensional isotope approaches, particularly the study of mass-independent fractionation, has transformed CAS from a basic geochemical proxy into a powerful, multifaceted tool for reconstructing paleo-atmospheric and oceanic redox conditions. This review synthesizes current understanding of CAS formation mechanisms, the challenges of post-depositional alteration and preservation, and the analytical innovations reshaping the field. By combining rigorous sequential extraction protocols with multi-isotope analyses, researchers can now robustly separate primary marine signals from diagenetic overprints and modern contamination. These methodological and conceptual advances have driven three fundamental transitions in CAS research: (1) from sparse, isolated data points to high-resolution chemostratigraphic records; (2) from single-isotope proxies to integrated multi-isotope frameworks; and (3) from largely qualitative interpretations to quantitatively constrained, process-based models. When synthesized with Earth-system modeling, these high-dimensional CAS datasets offer an increasingly precise window into Earth's oxygenation history, redox-climate feedbacks, and the long-term co-evolution of life and planetary habitability.