

FACIES MATTER: INTERPRETING BIOSIGNATURES IN ORGANIOSEDIMENTARY DEPOSITS FROM THE SALAR DE ATACAMA

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KEY FINDINGS

- Organosedimentary deposits (endoevaporites, evaporite sediments, carbonate sediments, sheet mats, discrete microbial buildups) record distinct stable carbon and sulfur isotope ratios.
- Mineralogical composition, spatial distribution of fractions, and microbial activity influence these geochemical signals.

INTRODUCTION

Since at least 3.5 billion years ago, microbes have influenced and/or induced the formation of sediments, leaving behind a long-standing archive of biological activity. Organosedimentary deposits can consist of mixtures of carbonate and evaporite minerals, as well as organic matter (Reid et al., 2024). Each of these components can serve as an archive for stable carbon and sulfur isotope ratios, the fractionation of which may be influenced by microbial processes (Thomazo et al., 2009). However, interpreting stable isotope ratios as chemical biosignatures is complicated by uncertainty about how changes in the proportion, spatial distribution, and mineralogy of the fractions comprising organosedimentary deposits influence the fractionation of stable carbon ($\delta^{13}\text{C}$) and sulfur ($\delta^{34}\text{S}$) isotope ratios.

DATA SET AND METHODS

We investigated modern sedimentary deposits from saline lakes (locally termed lagoons) along the eastern margin of the Salar de Atacama (Northern Chile), where endoevaporites, evaporite sediments, carbonate sediments, sheet mats, and discrete microbial buildups coexist (Fig. 1). From these facies, we collected a total of 101 lake bottom type samples. Sedimentological characteristics were documented through thin sections and petrography, SEM imaging, and XRD analyses. Geochemical measurements included $\delta^{13}\text{C}$ values of organic matter ($\delta^{13}\text{C}_{\text{org}}$) and carbonate minerals ($\delta^{13}\text{C}_{\text{carb}}$), and $\delta^{34}\text{S}$ values of organic matter ($\delta^{34}\text{S}_{\text{org}}$), measured using a Thermo EA IsoLink CNS + Delta Q IRMS. In addition, we conducted the first measurements of the $\delta^{34}\text{S}$ values of gypsum ($\delta^{34}\text{S}_{\text{gypsum}}$) using an Agilent 8900 ICP-QQQ upgraded with axial acceleration. To contextualize these biogeochemical datasets, we analyzed the corresponding lake waters associated with each lake bottom type sample, measuring the $\delta^{13}\text{C}$ values of dissolved inorganic carbon ($\delta^{13}\text{C}_{\text{DIC}}$) and $\delta^{34}\text{S}$ values of dissolved sulfate ($\delta^{34}\text{S}_{\text{SO}_4}$). These combined measurements enabled the calculation of apparent

isotope fractionation factors (Δ -values) between each organosedimentary facies and the overlying water column, as follows:

$$\Delta^{13}C_{\text{Carb-DIC}} = \delta^{13}C_{\text{carb}} - \delta^{13}C_{\text{DIC}}$$

$$\Delta^{34}S_{\text{Gypsum-SO}_4} = \delta^{34}S_{\text{Gypsum}} - \delta^{34}S_{\text{SO}_4}$$

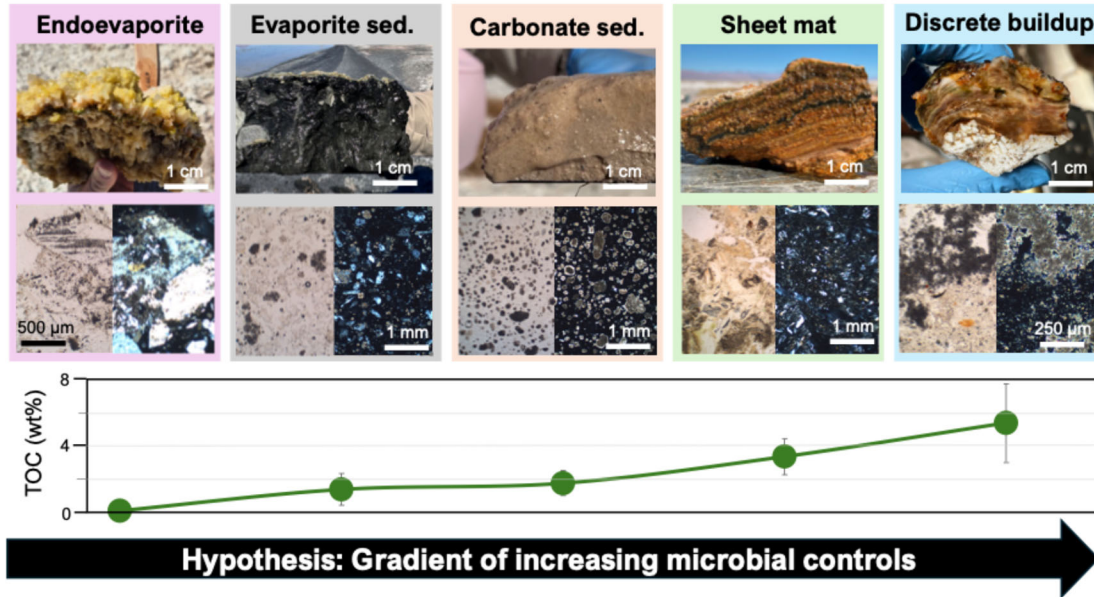


Figure 1. From top to bottom: field photographs, thin section images (natural light on the left, polarized light on the right), and total organic carbon (TOC, wt%) of five organosedimentary facies identified in the Soncor and Aguas de Quelana lake systems of the Salar de Atacama: endoevaporites, evaporite sediments, carbonate sediments, sheet mats, and discrete microbial buildups.

RESULTS AND INTERPRETATION

Sedimentology

Field observations, petrographic descriptions, and TOC measurements allowed us to distinguish five categories of organosedimentary deposits in the Salar de Atacama (Fig. 1). Endoevaporites are composed of large gypsum crystals with very low organic matter content (0.1 ± 0.1 wt% TOC), mostly trapped between or within crystals. Evaporite sediments consist of dark, fine-grained, gypsum-rich sediments containing more abundant organic matter (1.4 ± 1.0 wt% TOC) but lacking visible sedimentological stratification or microbial laminations. Carbonate sediments are typically beige to pink colored, dominated by micrite and peloids, intermixed with organic matter and similarly present as unlaminated deposits. Sheet mats form laterally extensive, finely laminated organic-rich layers (3.4 ± 1.1 wt% TOC), locally containing authigenic carbonate grains or thin lithified horizons. Discrete microbial buildups are three-dimensional microbial structures characterized by relief above the lakebed, well-developed alternations of organic-rich and lithified laminae, with a carbonate-dominated mineralogy (5.4 ± 2.4 wt% TOC). Taken together, these facies are interpreted to define a gradient of increasing microbial control on sediment chemistry, mineralogy, and architecture: from low-TOC evaporites dominated by crystalline gypsum, through muds with higher organic matter and authigenic micrite, to stratified mats and

fully developed microbial buildups (Fig. 1). This facies framework provided the basis for testing whether and to what extent stable isotope ratios reflect microbial activity and thus serve as biosignatures along this gradient.

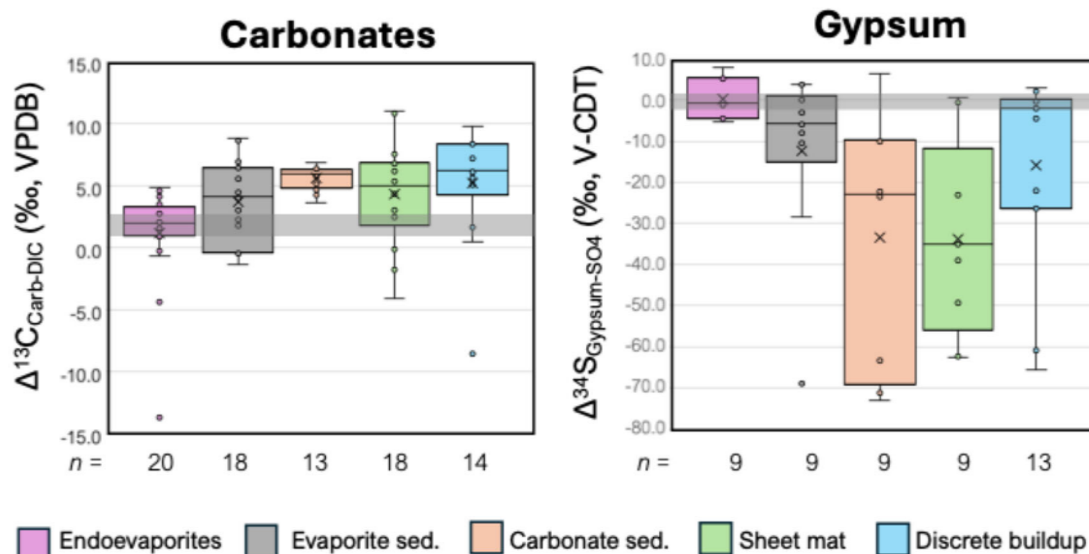


Figure 1. $\Delta^{13}\text{C}_{\text{carb-DIC}}$ (left) and $\Delta^{34}\text{S}_{\text{gypsum-SO}_4}$ (right) values for the five organosedimentary facies shown in Figure 1. The horizontal grey bar indicates the range of theoretical equilibrium isotope fractionation factors expected between carbonate minerals of various mineralogies and bicarbonate (Rubinson and Clayton, 1969; Romanek et al. 1992), and between gypsum and dissolved sulfate (Holser and Kaplan, 1966), providing an inorganic baseline for assessing isotopic disequilibria attributable to microbial processes.

Stable Isotope Geochemistry

Calculation of $\Delta^{13}\text{C}_{\text{carb-DIC}}$ values indicated pronounced differences among organosediment types (Fig. 2), ranging from $+1.1 \pm 4.1$ ‰ in endoevaporites to $+5.2 \pm 4.7$ ‰ in discrete microbial buildups, with discrete microbial buildups reaching $\Delta^{13}\text{C}_{\text{carb-DIC}}$ values as high as $+11.1$ ‰ V-PDB. Likewise, $\Delta^{34}\text{S}_{\text{gypsum-SO}_4}$ values varied widely (Fig. 2), from $+0.2 \pm 4.9$ ‰ in endoevaporite deposits to -33.9 ± 4.5 ‰ in sheet mats, with the lowest values reaching -73.1 ‰ V-CDT. When compared with the expected equilibrium isotope fractionations between water and the corresponding minerals (carbonate–DIC, Rubinson and Clayton, 1969; Romanek et al. 1992; and gypsum–dissolved sulfate; Holser and Kaplan, 1966), these results indicate a systematic departure from equilibrium conditions along the facies gradient shown in Figure 1. Minerals associated with endoevaporite deposits display $\Delta^{13}\text{C}_{\text{carb-DIC}}$ and $\Delta^{34}\text{S}_{\text{gypsum-SO}_4}$ values close to predicted isotopic equilibrium, consistent with predominantly abiotic precipitation mechanisms, whereas evaporite and carbonate sediments, sheet mats, and microbial buildups exhibit progressively larger offsets (Fig. 2). These trends are interpreted to reflect increasing degrees of microbial effects on carbon and sulfur cycling within the microenvironments in which these minerals form.

Implications

Results demonstrate that interpretation of stable isotope values is inherently facies-dependent. In endoevaporite deposits, $\Delta^{13}\text{C}_{\text{carb-DIC}}$ and $\Delta^{34}\text{S}_{\text{Gypsum-SO}_4}$ values plot near expected equilibrium values based on known mineralogical fractionation factors and lake water isotopic composition, indicating limited microbial influence on their stable isotope geochemistry. By contrast, $\Delta^{13}\text{C}_{\text{carb-DIC}}$ and $\Delta^{34}\text{S}_{\text{Gypsum-SO}_4}$ values calculated for sheet mats and discrete microbial buildups exhibit large departures from equilibrium predictions, consistent with substantial effects from microbial activity and high probability to record chemical biosignatures. Notably, $\Delta^{13}\text{C}_{\text{carb-DIC}}$ and $\Delta^{34}\text{S}_{\text{Gypsum-SO}_4}$ values in carbonate and evaporite sediments also exhibit significant isotopic disequilibria, in some cases overlapping with the values calculated for sheet mats, showing facies lacking morphological evidence of microbial activity can preserve robust chemical biosignatures. Collectively, endoevaporites, evaporite and carbonate muds, sheet mats, and discrete buildups each capture unique information about environmental conditions and microbe–mineral interactions. To integrate these facies-dependent signals, we propose a conceptual model that compares geochemical datasets across facies, providing multiple independent lines of evidence for reconstructing the degree of microbial activity and the formation of chemical biosignatures. Because diverse organosedimentary facies commonly co-occur, this model enables chemical biosignatures to be tracked continuously through stratigraphic successions, rather than inferred from a single facies. Advancing our knowledge of organosediment formation in the Salar de Atacama can refine conceptual models for ancient microbial carbonate deposits in analogous lacustrine systems (i.e., Virgone et al., 2013).

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