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Non-traditional biogeochemical redox cascades in extensional basins of the Cristiana-Santorini-Kolumbo Volcanic Field

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Microbial metabolisms in marine sediments typically follow the classical redox cascade based on the energy yielded from each metabolic reaction. In many marine depositional environments, the terminal electron acceptors for these metabolisms (e.g., sulfate or iron oxides) are supplied by diffusion from seawater (sulfate) or primary minerals (iron oxide particles). This sequence of reactions, from aerobic respiration to sulfate reduction and methanogenesis, impacts elemental concentrations, isotopes, and speciation, often overprinting primary signals with diagenetic signatures in relatively predictable ways. In tectonically and volcanically active environments, however, non-traditional depositional pathways and histories can upend this redox cascade, creating dynamic conditions that could be both detrimental and beneficial for microorganisms in deep marine sediments while adding complexity to interpretations of sedimentary biogeochemical archives.

The Hellenic Volcanic Arc in the Eastern Mediterranean hosts several active volcanic centers, including the Cristiana-Santorini-Kolumbo Volcanic Field (CSKVF). International Ocean Discovery Program (IODP) Expedition 398 drilled within the Santorini caldera and surrounding extensional basins, recovering sediments and volcanics to characterize the formation and eruptive history of the CSKVF and impacts of volcanism on the deep biosphere. A suite of porewater analyses were performed and identified complex, non-traditional behaviors of a variety of porewater constituents, including salinity, major cations (e.g., Ca^{2+}) and anions (e.g., SO_4^{2-}), and minor and trace elements. Isotopic analyses of water isotopes ($\partial^{18}O$ and $\partial^{2}H$), sulfur isotopes ($\partial^{34}S\text{-}SO_4^{2-}$), and strontium isotopes ($^{87/86}Sr$) reveal multiple diagenetic controls on porewater geochemistry, including contributions from brines originating in Messinian evaporites, volcanic ash alteration, bacterial sulfate reduction, and authigenic carbonate formation. Sediments and porewaters include up to four sulfate-methane transitions, with sulfate resupplied by vertical and lateral fluid migration along pathways controlled by the complex volcano-tectonic evolution of the basins and alteration of sedimentary physical properties during rapid deposition of erupted volcanics.

Microorganisms in CSKVF basins are continuously affected by changing metal and substrate availability with significant spatiotemporal heterogeneity. Our integrated isotopic investigation reveals non-traditional metabolic pathways and carbon cycling in active volcano-tectonic environments. The reactivated deep biosphere, supported by substrates resupplied by subsurface fluid flow, results in geochemical overprinting and a record of complex depositional and diagenetic regimes.