学 术 报 告

Uptake of metal cofactors in enzymes as a fingerprint for deep-time microbial C-fixation—Lessons learned from the Cd isotope system in Western Australian stromatolites



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报告简介：

Microbial habitats record the intricate interactions of the biosphere, atmosphere, and hydrosphere. The evolution of C-fixation in microbial ecosystems is directly linked to their metallome, i.e., the availability of dissolved metals to be used as enzymatic cofactors for diverse metabolism. One of the key metalloenzymes in the microbial world is carbonic anhydrase (CA), which plays an essential role in the carbon cycle of aqueous ecosystems by regulating the balance of CO_2 and HCO_3^-. In a microbial mat, diurnal fluctuation in light penetration causes steep chemical gradients and variations in gas exchange between variable microbes and ambient pore fluids on a millimeter scale. While phototrophic microbes in the upper layers create favorable conditions for authigenic carbonate precipitation, fermentation in the deeper layers causes partial carbonate dissolution, necessitating efficient carbon utilization across diverse microbial communities. Carbonic anhydrase is a cambialistic enzyme that commonly uses Zn in modern aerobic phototrophs. However, in a Zn-limited niche, Cd^{2+} is relatively more accessible. The operational use of Cd as an alternative cofactor in CA is mechanistically linked to the specific environmental conditions of microbial habitats, i.e., restricted, evaporative settings with high salinity, high pH, and localized sulfide formation via bacterial sulfate reduction. Stable Cd isotope fractionation during CA-uptake by modern phototrophs is readily recorded in authigenic carbonates. We test whether this proxy can be expanded to trace CA in the microbial rock record. We analyzed Cd isotopic compositions in stromatolite occurrences spanning the Archean to the Holocene and report fractionated Cd isotopic compositions that correlate with macro- and micronutrients P, Cd, and Zn, suggesting enzymatic metal cycling from the Paleoarchean onwards. Meanwhile, Rayleigh-type Cd isotope fractionation indicates its use as a cofactor from as early as the Meso- to Neoarchean, with long-term evolution in Cd/Zn ratios and Cd isotopic compositions approaching modern surface-ocean values before the GOE. We further show that the temporal restriction of a continental shelf during the Badenian Salinity Crisis in the Miocene is recorded in the Cd isotope compositions of stromatolitic carbonates deposited in lagoon-like conditions. Our work on Cd isotope compositions in stromatolites offers new insights into the metallome and its role in facilitating microbial habitats, adding a new isotope-biomarker perspective to the study of deep-time microbial evolution.