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Uniquely low stable iron isotopic signatures in deep marine sediments caused by Rayleigh distillation

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Microbially mediated iron (Fe) reduction is suggested to be one of the earliest metabolic pathways on Earth and Fe(III)-reducing microorganisms might be key inhabitants of the deep and hot biosphere [1, 2]. Since microbial Fe cycling is typically accompanied by Fe isotope fractionation, stable Fe isotopes (δ^{56}Fe) are used as tracer for microbial processes in modern and ancient marine sediments [3, 4]. Here we present Fe isotope data for dissolved and sequentially extracted sedimentary Fe pools from subseafloor sediments that were recovered during International Ocean Discovery Program Expedition 370 from a 1,180 m deep hole drilled in the Nankai Trough off Japan where temperatures of up to 120°C are reached at the sediment-basement interface. The expedition aimed at exploring the temperature limit of microbial life and identifying geochemical and microbial signatures that differentiate the biotic and abiotic realms [5, 6]. Dissolved Fe ($Fe(II)_{aq}$) is isotopically light throughout the ferruginous sediment interval but some samples have exceptionally light δ^{56}Fe values. Such light δ^{56}Fe values have never been reported in natural marine environments and cannot be solely attributed to microbially mediated Fe(III) reduction. We show that the light δ^{56}Fe values are best explained by a Rayleigh distillation model where $Fe(II)_{aq}$ is continuously removed from the pore water by diffusion and adsorption onto Fe (oxyhydr)oxide surfaces. While the microbially mediated $Fe(II)_{aq}$ release has ceased due to an increase in temperature beyond the threshold of mesophilic microorganisms, the abiotic diffusional and adsorptive $Fe(II)_{aq}$ removal continued, leading to uniquely light δ^{56}Fe values. These findings have important implications for the interpretation of Fe isotope records especially in deep subseafloor sediments.

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