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Extraordinary ^{13}C-enrichment of DIC produced by acetoclastic methanogen-dominated microbial consortia

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Extreme carbon isotopic signatures have long been associated with the methane and biomass produced by methanogens[1]. Large fractionations are associated with hydrogenotrophic methanogenesis, while methylotrophic and acetoclastic methanogenesis typically produce less pronounced ^{13}C-fractionations[2,3]. Isotopic signatures of inorganic carbon in methanogenic systems have received less attention. ^{13}C-enrichment of coalbed CO_2 has been attributed to drawdown by methanogens balanced by production via respiration[4]. Yet, large ^{13}C-enrichments in dissolved inorganic carbon (DIC) have been observed in petroleum formation waters and aquifers with no apparent distillation[5,6]. Here we report on and evaluate extraordinary ^{13}C-enrichment and accumulation of DIC in petroleum reservoirs within the Uinta Basin, USA, integrating geomicrobiology, geochemistry, and hydrogeology. We find adjacent reservoirs are discrete subsurface ecosystems with complex ecology influenced by salinity, temperature, permeability, and geologic history. Strongly ^{13}C-enriched DIC is found in sites dominated by the acetoclastic methanogen Methanosaeta with low dissolved acetate (Figure 1). Network analysis reveals tight and consistent associations between Methanosaeta, fermenters (namely JS1), diverse sulfate reducers, as well as hydrogenotrophic methanogens. In seeking a mechanism to explain both DIC accumulation and ^{13}C enrichment, we evaluate the anaerobic food chain. Organic acids, especially acetate, in petroleum reservoirs are ^{13}C-enriched relative to bulk oil, even when temperature exceeds habitability, and amplified where temperatures are lower[5,7]. Pure culture studies confirm strong ^{13}C-enrichments of acetate during fermentation[8]. We suggest that the observed ^{13}C-signatures develop through time, beginning with organic matter biosynthesis predating oil formation, then via fermentation reactions, and finally distillation of the dissolved acetate pool. An anerobic food chain based in fermentation, utilization of organic acids, then cleavage of enriched acetate by acetoclastic methanogens provides a stepwise mechanism to transfer ^{13}C-enriched organic carbon into the inorganic pool, while increasing alkalinity. Our documentation of highly ^{13}C-enriched carbonate minerals in the Paradox basin, and as observed elsewhere[9] provide a mechanism to preserve this biosignature in the geological record.

1. Whiticar+, GCA (1986); 2. Gelwicks+, AEM (1994); 3. Gropp+ Sci.Adv. (2022); 4. Vinson+ Chem.Geol. (2017); 5. Carothers & Kharaka, GCA (1980); 6. Chapelle & Knobel, Groundwater (1985); 7. Franks+ GCA (2001); 8. Mueller+ PNAS (2025); 9. Liu+ Geology (2024).

