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Radiolysis-Driven Evolution of an Ancient Subsurface Habitable Brine in the Witwatersrand Basin, South Africa

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Crustal fluids can evolve elevated salinities over Ma-Ga timescales via extended water-rock interactions, with hydrogeochemical history reflected in their geochemical and isotopic compositions [1][2]. In radionuclide-enriched subsurface fractures, the decay of ^{238}U, ^{232}Th and ^{40}K triggers radiolysis of water, and drives increasing salinity and the formation of redox and organic species fueling inhabiting biota [3]. We present a case of alpha-particle radiolysis driven brine formation and support for microbial metabolism in a deep (3.2 km), ancient (1.2 Ga) and thermal (45-54 °C) subsurface brine in Moab Khotsong gold and uranium mine of the Witwatersrand Basin, South Africa [4][5]. This brine (215-246 g/L TDS) was found to have the highest radiolytically produced excess of ^{86}Kr and a ^{136}Xe excess consistent with uranium fission [5]. Estimated concentrations of ^{238}U for this system (1-100 ppm) account for local annual dosages of 0.02–0.3 Gy and minimum estimates of annual redox species production for H_2 (7 nM), NO_2^- (2 nM), NO_3^- (0.1nM), SO_4^{2-} (0.2 nM), and O_2 (3 nM) [4]. Introduction of redox species via recent mixing with meteoric waters or air exchange were not supported, based on $\Delta^{14}C$ dating and non-meteoric $^{18}O/^{2}H$ isotopic signatures for this system, suggesting their autochthonous origin. Additional analyses of stable isotopes $\delta^{18}O_{calcite}$, $\delta^{13}C_{calcite}$, $\Delta^{33}S_{pyrite}$, $\delta^{34}S_{pyrite}$ and $^{87}Sr/^{86}Sr$ confirm the interaction of multiple past fluid incursions with surrounding strata [4]. The brine currently contains a low biomass bacterial community (10^2-10^4 cells/mL) with favored metabolic strategies including aerobic heterotrophic, fermentative, denitrifying and thiosulfate oxidizing members. The Moab brine system represents a high salinity and ancient end member distinct from less saline paleo-meteoric fluids in the Basin [6]. These findings provide a new opportunity to examine radiolytic-driven brine formation in an ancient subsurface brine system with abiotic support for a low biomass biosphere. [1] Drake et al. (2021) *Commun. Earth Environ*. 2, 102 [2] Warr et al. (2021) *Chem. Geol.* 561, 120027 [3] Sherwood Lollar et al. (2021) *Geochim. Cosmochim. Acta* 294, 295-314. [4] Nisson et al. (2023) *Geochim. Cosmochim. Acta* 340, 65-84. [5] Warr et al. (2022) *Nat. Comms.* 13, 3768-3768 [6] Onstott et al. (2006) *Geomicrobiol. J.* 23, 369-414.