

Emergence of oxygenic photosynthesis and the probability of natural fission reactors: A negative feedback to explain the delayed oxidation of Earth's surface?

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The rise of free oxygen in Earth's atmosphere resulted from the proliferation of the photosynthetic cyanobacteria. Fossil and molecular biomarker data from the geologic record date the origin of the cyanobacteria to 2.7 Gyr if not earlier. Evidence suggests the transition from an initial, virtually anoxic atmosphere to one with persistent free oxygen occurred as late as 2.4 Gyr ago leaving a significant lag between the emergence of oxygenic photosynthesis and the irreversible oxidation of the Earth's surface. Explanations for this delay commonly suggest secular changes in the balance between the fluxes of oxygen and reducing equivalents to the atmosphere coincident with the ~2.4 Gyr transition. Models include timely increases in the burial of organic matter, a decline in the content of reducing equivalents in volcanic and metamorphic source gases and progressive methane mediated hydrogen escape. Here we present calculations supporting the idea that due to its redox sensitivity, uranium deposits should have formed in the isolated marine or freshwater environments where oxygenic photosynthetic organisms first took hold and established strong local reduction-oxidation gradients. These are predicted to have formed near-surface critical natural fission reactors. Natural fission reactors represent point sources of radiolytic energy that would result in the local release of heat, ionizing radiation and free radicals and farfield effects through the production of mobile short- and long-lived radioactive daughter isotopes. We discuss the speculative scenario that the association of critical actinide deposits with photosynthetic point sources of oxygen could provide a negative feedback limiting the proliferation of the cyanobacteria in the Archean environment. Secular decreases in the ²³⁵U/²³⁸U ratio would undoubtedly decrease the probability of critical deposits through time reducing the impact of this feedback.

Reactive transport modeling of U(VI) in small-scale tracer tests

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Two small-scale tracer tests were conducted to evaluate the applicability of a semi-mechanistic U(VI) adsorption model developed in the laboratory for describing the impact of variable alkalinity and Ca on reactive transport in the field. The experiments were conducted in a region of an alluvial aquifer located beneath a former uranium mill near Naturita, CO that had a U(VI) concentration equal to 4 μM, an alkalinity equal to 8 meq/L and a pH of 7.1. The ambient groundwater was close to equilibrium with calcite (0.0 < SI < 0.1). The experiments were conducted by extracting contaminated groundwater from the aquifer and adjusting the alkalinity to either 25 meq/L by adding sodium bicarbonate or 2.5 meq/L by adding hydrochloric acid. In both experiments 500 mg/L Br was added as KBr to serve as a nonreactive tracer and the pH was held constant by maintaining a constant partial pressure of CO₂. The treated groundwater was injected into a line of three wells and breakthrough was monitored at four multilevel wells located 1.8m and 2.4m downgradient of the injection wells.

Previous studies have shown that the transport of U(VI) is sensitive to alkalinity and that the dominant species in solution are Ca₂UO₂(CO₃)₃⁰ and CaUO₂(CO₃)₃⁻² suggesting that Ca also can significantly affect U(VI) transport. For the tracer tests where the alkalinity was increased to 25 meq/L, the additional carbonate caused the groundwater to become oversaturated with respect to calcite (SI = 0.4). The extent of oversaturation of calcite increased during the tracer test to saturation indices as large as 0.65 suggesting that calcite did not precipitate during the experiment. In addition, the temporal changes in Ca, Mg, and Na suggested that ion exchange reactions driven by increased Na and K concentrations were important. The U(VI) concentrations in the monitoring wells initially increased to 12 μM, then decreased to 2.5 μM and finally returned to the initial concentration of 4 μM. The observed U(VI), alkalinity, Ca, Mg and Na concentrations were reproduced using a reactive transport model incorporating the semi-mechanistic U(VI) surface complexation model, ion exchange reactions, and kinetically-controlled calcite precipitation and dissolution. Analyses of the second tracer test where the alkalinity was decreased are currently being conducted.