

Anoxic photochemical weathering of pyrite on Archean continents

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Background: On the early Earth, pyrite weathering by atmospheric oxygen was severely limited and low marine sulfate concentrations persisted for much of the Archean eon. Despite the anoxic conditions, an increase in the total sulfur weathering flux and the mobilization of transition metals from sulfide minerals have been observed in marine sedimentary records, suggesting that sulfide mineral oxidation on land occurred prior to the Great Oxidation Event. Here we show an anoxic photochemical mechanism of pyrite weathering that could have provided significant amounts of sulfate to the oceans as continents formed in the late Archean.

Materials and Methods: Anoxic UV irradiation experiments were conducted by mixing pyrite grains with a ferrous chloride solution in N₂ purged quartz reaction vessels. A 450-watt Hg vapor lamp in a photochemical quartz immersion well was used to irradiate the pyrite suspensions. At periodic intervals, the concentration of dissolved sulfate was measured using ion chromatography. To investigate pyrite weathering in the late Archean, UV irradiation experiments were conducted with 2.68 Ga pyritic shale collected at the Golden Mile Super Pit, Western Australia.

Results and Discussion: Pyrite grains suspended in anoxic ferrous iron solutions produced millimolar levels of sulfate when irradiated with UV light. The Fe²⁺_(aq) was photooxidized, which in turn, led to the chemical oxidation of pyritic sulfur. Experiments conducted with 2.68 Ga shale demonstrated that photochemically derived ferric iron oxidizes and dissolves sedimentary pyrite during chemical weathering. A photogeochemical model was developed to constrain the rates of sulfate production from pyrite oxidation by photochemically derived Fe³⁺. The results suggest that prior to the rise of atmospheric oxygen, oxidative pyrite weathering on Archean continents was controlled by the exposure of land to sunlight.