Pyrite oxidation initiates weathering reactions in shale (and other lithologies?) at depth

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The deepest weathering reaction we have identified under the Susquehanna Shale Hills Critical Zone Observatory is pyrite oxidation. Cuttings of Rose Hill shale were investigated from one borehole from the ridge and four boreholes from the valley at the CZO. Pyrite concentrations are insignificant to depths of 23 m under the ridge and 8–9 m under the valley. Likewise, carbonate concentrations are insignificant to 22 and 2 m, respectively. In addition, a 5–6 m-thick fractured layer directly beneath the land surface shows evidence for loss of illite, chlorite, and feldspar. Under the valley, secondary carbonates may have precipited. The limited number of boreholes and the tight folding make it impossible to prove that depth variations result from weathering instead of chemical heterogeneity within the parent shale. However, carbonate depletion coincides with the winter water table observed at ~20 m (ridge) and ~2 m depth (valley). It would be fortuitous if carbonate-containing strata are found under ridge and valley only beneath the water table. Furthermore, pyrite and carbonate react quickly and many deep reaction fronts for these minerals are described in the literature. We propose that deep transport of O_2 initiates weathering at SSHO and many other localities because pyrite commonly oxidizes autocatalytically to acidify porewaters and open porosity. According to this hypothesis, the mineral distributions at SSHO are nested reaction fronts that overprint protolith stratigraphy. The fronts are hypothesized to lie subparallel to the land surface because O_2 diffuses to the water table and causes oxidative dissolution of pyrite. Pyrite-derived sulfuric acid (H_2SO_4) plus CO_2 also dissolve carbonates above the water table. To understand how reaction fronts record long-term coupling between erosion and weathering will require intensive mapping of the subsurface.