Depth variation of soil iron crystallinity at the Calhoun Critical Zone Observatory

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The crystallinity of iron minerals can determine the impact iron has on ecosystem functions. Soil iron serves three broad categorical functions in terrestrial ecosystems: (1) as a structural component of the soil matrix and as a cementing bridge in soil aggregates; (2) as a sorbent retaining P, C and trace elements; and (3) as a host to electron-transfer reactions including acting as a terminal electron acceptor during anaerobic soil respiration. The degree of Fe phase crystallinity has great significance because it is typically inversely related to mineral surface area and by extension adsorptive (2) and electron transfer (3) reactivity.

As a consequence of pedogenic processes, we hypothesized that Fe phase crystallinity would increase with soil depth, yielding an greater abundance of short-range-ordered Fe phases in the surface horizons where iron likely plays a larger role in ecosystem function. We tested this hypothesis using a well characterized profile of alternating Fe enriched and Fe depleted microsites (intercalated light (yellow/white) and dark (red) stripes) at the Calhoun Critical Zone Observatory (CZO) in South Carolina, USA. We sampled these microsites from 56 to 183 cm depth, and also sample the surface (0-15 cm) and subsurface horizons (15-56 cm), which were more homogeneous in terms of Fe abundance. We characterized these redoxmorphic features via total elemental analysis, X-ray diffraction (XRD) and ^{57}Fe Mössbauer spectroscopy (MBS). Concentrations of Si, Al, Ca, and Mn did not differ between Fe-enriched and Fe-depleted zones, which contained 2.6 - 6.1% and 1.5 - 2.3% Fe, respectively. The Mössbauer spectra clearly indicate hematite and goethite in the Fe-enriched spectra, with crystallinity increasing with depth, in support of our hypothesis. The Fe-depleted microsites contained Fe primarily in clay minerals (e.g., kaolinte and other layered silicates), but became enriched in Fe (oxyhydr)oxides closer to the surface. This variation in Fe phase crystallinity within similar redoxomorphic features suggests the role of surface processes (i.e., vegetation) can influence soil development well below the typical rooting zone.