The association of organic matter (OM) with minerals has been increasingly recognized as a major process for protecting organic matter against microbial degradation in soils. Iron (Fe) oxides are of particular importance because of their abundance in soils and high reactive surface area. Iron is susceptible to redox variability along landscape gradients. Reductive dissolution and transformation of Fe minerals governs the amount, form and transport of sequestered C. In this study, we investigated the Fe speciation as well as the composition of organic matter and its molecular interaction with soil minerals across hillslope transects within the Christina River Basin Critical Zone Observatory (CRB-CZO) to link iron-redox coupling processes with soil C cycling. Selective chemical extractions, X-ray absorption spectroscopy (XAS) and micro-XAS techniques were used to characterize soil Fe species. Applying scanning transmission X-ray microscopy and near edge X-ray absorption fine structure spectroscopy (STXM/NEXAFS), we mapped the spatial distribution of carbon and carbon forms, and imaged the association of organic functional groups with specific minerals in the soils. We used both C-NEXAFS and pyrolysis-field ionization mass spectrometry (Py-FIMS) to speciate the organic matter at the molecular scale. Furthermore, Py-FIMS unequivocally showed that the thermal stability of specific organic compound classes in the soils was affected by association with Fe oxides. This investigation will enhance our understanding of OM stabilization mechanisms on soil mineral surfaces and will provide new insights on the Fe-redox coupling processes affecting carbon cycling at soil/sediment-water interfaces.