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Iron Redox Dynamics Across Coastal Terrestrial-Aquatic Interfaces: Field Study in The Great Lakes and Chesapeake Bay regions

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Terrestrial-aquatic interfaces are dynamic environments with spatial and seasonal redox fluctuations that impact the geochemical behavior of redox sensitive elements such as iron (Fe) and sulfur (S), as well as the biogeochemical cycling of carbon, nutrients, and contaminants [1,2]. The Coastal Observations, Mechanisms, and Predictions Across Systems and Scales-Field, Measurements, and Experiments (COMPASS-FME) pilot study aims to better understand the mechanisms that control the structure, function, and evolution of coastal terrestrial-aquatic ecosystems. As part of this project our study aims to better understand Fe redox biogeochemistry across coastal sites in the Great Lakes and the Mid-Atlantic regions of the United States.

Using X-ray Absorption Spectroscopy, the oxidation state and molecular environment of Fe was determined with depth in undisturbed soil cores collected across upland to shoreline gradients from coastal locations along Lake Erie (freshwater) and Chesapeake Bay (marine). X-ray absorption near edge spectroscopy results show that Fe occurs mainly in its oxidized form, Fe(III), in cores collected from upland locations in both the Lake Erie and Chesapeake Bay regions. In soil cores collected closer to the shoreline (i.e., forest transition zones and coastal wetlands), Fe is partitioned between Fe(III) and Fe(II), with Fe(II) proportions increasing from the intermittently flooded to the permanently flooded locations. Extended x-ray absorption fine structure spectroscopy suggests that Fe(III) occurs as Fe-(hydr)oxide and Fe-bearing phyllosilicate minerals in both the Lake Erie and Chesapeake Bay regions. In contrast, Fe(II) speciation differs between Lake Erie and Chesapeake Bay. We observed the presence of Fe sulfides in the Chesapeake Bay wetland soils, indicating a sulfur-driven redox chemistry, whereas Fe(II) is mainly observed in Fe-bearing phyllosilicate minerals in the samples collected at Lake Erie sites. Complemented with mineralogical, solid and pore water chemistry information, these results contribute to a better understanding of Fe redox cycling in marine and freshwater coastal systems.

[1] Ward et al. (2020), *Nature Communications* 11,1:2458.
[2] Kappler et al. (2021) *Nature Reviews Microbiology* 19, 6.