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Biogeochemical Controls on Mobilization of Labile Carbon from Wood in Denitrifying Bioreactors: Insights from Multi-Modal Synchrotron Analysis of Wood Degradation

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Woodchips and other woody residues are commonly used as renewable and low-cost carbon sources and biofilm support structures in subsurface biofilter systems used to enhance removal of nitrate in stormwaters via microbial denitrification, a heterotrophic process coupling organic carbon oxidation with nitrate reduction. However, denitrification in these systems is typically carbon-limited due to the recalcitrance of lignin-rich structures in wood. The objective of this study is to understand the biogeochemical pathways behind the degradation of wood in redox-dynamic environments. There is a specific focus on understanding how the distribution of redox-sensitive metals (e.g., iron (Fe) and manganese (Mn)), affect the mobilization of bioavailable carbon from woody biomass. These redox reactions can be direct, like those on Mn(III/IV) oxide surfaces or with fungal Mn peroxidase(MnP) enzymes, or indirect via reactive oxygen species produced by Fe(II) oxidation.

We excavated woodchips from nine different locations in a subsurface denitrifying biofilter system that had been weathering in situ for more than 10 years, and characterized the woodchip with a range of bulk (C/N ratios; surface metal, biomass concentration and concentration of hydroxyl radicals) and micro-scale synchrotron-based techniques. Using X-ray scattering-based scanning tomography technique, we visualized the cellulose crystallinity index (C.I.) in woodchip specimens as a measure of degradation, and showed that wood that was exposed to oxygen was the most degraded. Higher concentrations of Fe and Mn on wood surfaces were also associated with greater levels of degradation, but it was unclear if the relationship is causal or due to the fact that degradation occurred at wood surfaces. Batch experiments showed that more degraded woodchips supported faster denitrification rate, likely due to greater biofilm concentrations in those samples resulting from greater carbon availability. The extraction of poorly-crystalline Fe and Mn oxides by pyrophosphate indicated that significant amounts of organic carbon were sequestered in minerals on woodchip surfaces, indicating that the bioavailability of carbon for denitrifies may be impacted. These results provide new insights into wood decomposition and labile carbon release in subsurface denitrifying biofilters, and highlight the complex roles that Fe and Mn oxide minerals can play in regulating carbon availability in these systems.