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An Integrated Method for Molecular Investigation of Soil Organic Carbon Composition, Variability, and Spatial Distribution Across a Sub-alpine Catchment

Hsiao-Tieh Hsu[1,2], Corey R. Lawrence[3], Matthew J. Winnick[1], and Kate Maher[1]

[1] Department of Geological Sciences, Stanford University, Stanford, CA
[2] Department of Chemistry, Stanford University, Stanford, CA
[3] US Geological Survey, Denver, CO

Contact: Hsiao-Tieh Hsu [hthsu9@stanford.edu] and Kate Maher [kmaher@stanford.edu]

Cycling of carbon through soils, the largest actively cycling terrestrial carbon reservoir, is one of the more poorly constrained components of the global carbon cycle. Although soil carbon models generally represent soil organic carbon (SOC) dynamics in terms of molecular labiality and recalcitrance, recent investigations suggest other processes, such as organicmineral associations and aggregate protection, may dominate SOC turnover rates, complicating responses to climate change. As a consequence, mechanistically based soil carbon models may produce results more aligned with observations of the carbon cycle. Constructing mechanistic model frameworks requires knowledge of SOC composition at a molecular-level, yet robust characterization of SOC compositions has proven challenging. Thus, in order to reduce the uncertainty in model predictions of the land-surface response to climate change, there is a pressing need to develop advanced methods for SOC characterization and to pair these strategies with development of new model frameworks.

I have developed a novel integrated methodology using Fourier transform infrared spectroscopy (FT-IR) and bulk carbon X-ray absorption spectroscopy (XAS) to identify SOC functional groups and elemental analysis (EA) to constrain the total available SOC so that we can study SOC composition quantitatively. I am responsible for IR, XAS and EA data collection, developing the algorithm for quantifying various SOC species, and using this method to study SOC variations across the sub-alpine catchment of East River, a shaledominated watershed located near Crested Butte, Colorado. I compared the differences in the amount of various SOC species between soils from two hillslope meadows located at different elevations and life zones (upper montane at Bradley Creek, and upper subalpine at Rock Creek) and a low-lying toe-slope soil dominated by willow (Bradley Creek). At Bradley Creek, polysaccharide, traditionally considered to be labile to microbial decomposition, is a significant portion of SOC while the aromatic class (*i.e*. aromatic, quinoic, and phenolic carbon), traditionally categorized as recalcitrant, is only present in a very small amount relative to litter inputs. At Rock Creek, a large amount of carbonyl and phenolic carbon is observed, possibly due to incomplete oxidation of other SOC species. Aliphatic carbon consistently makes up a significant proportion of SOC at all sites. Possible explanations for variations in SOC distribution between these sites include differences in plant inputs or microbial decomposition level. Additionally, I used scanning transmission X-ray microscopy (STXM) to map the spatial distribution of different SOC species on small soil particles. I found that aromatic carbon is mostly concentrated on the edge of soil particles, leading to larger areas of exposure and greater accessibility to microbes. On the other hand, polysaccharide, amide, and aliphatic carbons are spread throughout the soil particles, resulting in lower physical accessibility relative to their volume. We hypothesize that this difference in spatial distribution possibly explains the increase in labile SOC species relative to plant litter inputs. Our results further strengthen the shifting soil carbon paradigm that molecular structure and thermostability are subordinate to physical protection in controlling SOC turnover rates. Further, our newly developed characterization method provides, for the first time, the ability to link SOC quantities to reactivity and above ground ecosystem properties. This work uniquely allows us to integrate SOC speciation data into a new generation of soil carbon models that can better predict the land-surface response to climate change.