**Overview**

The dissolved fraction of organic carbon (DOC), transported by streams and rivers, plays an important role in the global carbon (C) cycle. Because forested headwater catchments cover large land areas, they have disproportionate effects on DOC dynamics and are closely monitored across the globe. Significant increases in DOC fluxes from forested streams across the northern hemisphere have been documented by numerous studies and potential causes such as changes in climate, land use or precipitation composition (i.e. recovery from acidification) are debated. The fact that little focus has been given to the precise mechanisms behind the DOC release makes the attribution of drivers difficult and the prediction of future DOC fluxes impossible. We propose to address this gap by combining modelling and experimental approaches to test the hypothese that 1) stream water DOC flux increase is driven by the regionally observed recovery from acidification (i.e. the increase in pH and decrease in ionic strength of wet and dry deposition), that 2) DOC is released from soil aggregates that become unstable under these changing conditions and that 3) aggregate stability and DOC release is a function soil composition and mineralogy, leading to the varied responses (presence or absence of DOC increase) despite potentially similar regional forcings. Big Data analysis using novel statistical modelling will be used on USGS and CZO datasets to probe regional scales (>100 km) and identify general patterns (testing hypotheses 1). Results from this step will inform selection of sites for more detailed process investigation at the catchment (km) to soil aggregate scale (micrometer) using Reactive Transport Modelling and experiments (testing hypotheses 2-3).

**Intellectual merit**

1) We tackle a highly debated topic in C dynamics (i.e. increase DOC fluxes) and furthermore work towards a template for the integration of scales, disciplines and approaches in low temperature geochemistry. The combination of statistical and process-based modelling with experiments to bridge scales varying >10 orders of magnitude is novel and potentially transformative for the field of low temperature geochemistry.

2) The use of these novel statistical tools on big data is currently leading to a fundamental change in experimental design where big data analyses is put at the front end of hypotheses development. A byproduct of this approach are less biased analyses that identify complex patterns and correlations which, in turn, lead to questions/hypotheses that have not yet been asked. As such, this research will provide additional findings that go beyond the scope of the proposed work and will be available to the geochemistry and Critical Zone community.

**Broader Impact**

1) We will provide interdisciplinary training in complex systems and the use of multiple approaches (ranging from field to lab experiments to modelling) for multiple graduate and undergraduate students.

2) Furthermore, we propose in collaboration with Dr. Regina Toolin (Associate Professor of Science Education in the College of Education at UVM) and the Sustainability Academy in Burlington, VT (minority serving K-5 elementary school), to develop and administer a professional development workshop for Vermont’s K-5 teachers. Goals for this workshop are to i) provide professional development for K-5 educators on the CZ as a framework for sustainability learning and ii) to begin to develop teaching modules with participating educators for the appropriate K-5 level. We will assess the effectiveness of the workshops with surveys at the end of each workshop day. Furthermore, teaching materials will be tested by faculty of the sustainability academy in their classroom and adapted as necessary before making them publicly available on the schools homepage. Even though the workshop will be held in Vermont, outreach by PIs Harpold and Li will aim for the dissemination of teaching materials to schools in their respective home states as well (NV and PA).