Quantifying regolith formation rates with U-series isotopes along the shale weathering transect within the Susquehanna Shale Hills Critical Zone Observatory

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Statement of Proposed Work

Regolith formation and chemical weathering are important Critical Zone processes. The generation of regolith from bedrock weathering releases nutrients to ecosystems, controls surface water chemistry, and maintains a soil cover at Earth's surface. The latter has become a major issue facing humanity as the loss of soil via erosion has increased greatly with human activities. It is important to understand rates and mechanisms of regolith formation to study sustainable soil development. Despite the fundamental importance, we still lack effective field tools to quantify the regolith formation process. More case studies to develop potential geochronologic tools to investigate regolith formation are in great need. U-series isotopes (e.g. ^{238}U, ^{234}U and ^{230}Th) have been recently utilized as a novel dating tool for Earth surface processes. This is largely due to improvements of analytical methods, mathematical approaches, and conceptual models made over the last decades in measuring U-series isotopes and interpreting their fractionation during chemical weathering.

Here, I propose to extend our current investigation of U-series isotopes at the Shale Hills catchment to the shale weathering transect established by the Shale Hills CZO team members Tim White, Ashlee Dere and Susan Brantley. I will focus on six small watersheds developed mainly on Fe-rich, organic-poor Silurian shale units along a latitudinal climate gradient in eastern United States and Wales. These field sites have been established within the Shale Hills CZO network to investigate the control of climates on soil chemistry and development. The proposed work will benefit significantly from the previous sampling campaigns carried out by the Shale Hills CZO team members. The new U-series isotope data set, in conjunction with the existing major and trace element data from these sites, will allow for detailed assessment of the U-series isotope fractionation processes and timescales of regolith formation on shale bedrock along the climate sequence. The quantification of regolith formation rates from shale bedrock will also help greatly to understand global element cycles, as shale bedrock covers up to ~30% of land surface and shale weathering contributes significantly to global fluxes of C, P, Pt- and REE group elements.

In our previous work at the Shale Hills catchment in central Pennsylvania, we measured U-series isotopes in six regolith profiles from two planar hillslopes to quantify regolith formation rates and to assess the role of hillslope aspect on regolith formation (Ma et al., 2010; and in review). All regolith samples displayed significant U-series isotope disequilibrium values due to shale weathering. These values showed systematic variations with regolith depths and were explained by a dual process of U-series isotope fractionation during weathering: a loss of ^{234}U, ^{238}U, and ^{230}Th during water-rock interactions and a gain from circulating soil water and/or downslope particle transport. With a U-series isotope mass balance model, regolith production rates (15 to 52 m/Ma) and weathering timescales (7 to 45 ka) were successfully calculated at Shale Hills. Systematical differences were observed for these two hillslope: the regolith profiles on the northern (sun-facing) slope are characterized by faster regolith production rates and shorter durations of chemical weathering in the regolith zone than those of the southern slope (shaded) but the ridge top sites have similar regolith production rates and durations of chemical weathering. These results reveal an important control of hillslope aspect on microclimatic conditions that in turn control the rate of regolith formation at Shale Hills. More importantly, our previous work highlights that the implementation of U-series isotopes to the Critical Zone studies has revolutionized our ability to evaluate the processes and timescales of regolith formation.

In collaboration with the Shale Hills CZO team members Tim White, Ashlee Dere and Susan Brantley, we will study soil profiles from small gray shale watersheds in Wales, New York, Virginia, Tennessee, Alabama, and Puerto Rico. Only ridge top sites were selected to limit the aspect effect. Mean annual air temperatures of these sites increase from 7 °C to 25 °C from north to south while annual precipitation ranges between 100 cm to 250 cm. These sites thus provide an ideal climo-sequence to study the effects of climate on gray shale weathering from cold and wet conditions to warm and wet conditions. The main goals of this project are:

1) To measure U-series isotope compositions in ridge top soil profiles along the shale weathering transect;
2) To understand the behavior of U-series isotopes during shale weathering and the controls of U-series disequilibrium signatures using the combined U-series isotope, major and trace element, and mineralogy data sets;
3) To determine regolith formation rates with the U-series disequilibrium signatures in a conceptual U-series mass balance model and to evaluate the effects of climate on rates of shale weathering.

In a pilot study, U-series isotopes have been measured in parts of the Virginia and Puerto Rico profiles at University of Texas at El Paso (UTEP). These preliminary results show U-series isotope disequilibrium as expected (Fig. 1). However, when compared to the Shale Hills profiles, these new profiles show more complex variation patterns of U-series disequilibrium with respect to depth. For example, in the shallow parts of these profiles (< 40 cm at Virginia and < 300 cm at Puerto Rico; Fig. 1), both ($^{234}U/^{238}U$) and ($^{230}Th/^{238}U$) values decrease systematically towards the surface, suggesting mobile behaviors for both ^{230}Th and ^{234}U isotopes. This is in contrast to the previous Shale Hills profiles in which ^{230}Th was observed to be much less mobile than ^{234}U and ^{238}U. More intensive leaching by organic acids in shallow soils might play a role in the observed ^{230}Th mobility. These systematic variations of U-series activity values in the shallow parts of these profiles can allow for calculation of regolith formation rates with a mass balance model similar to Ma et al. (2010; in review).

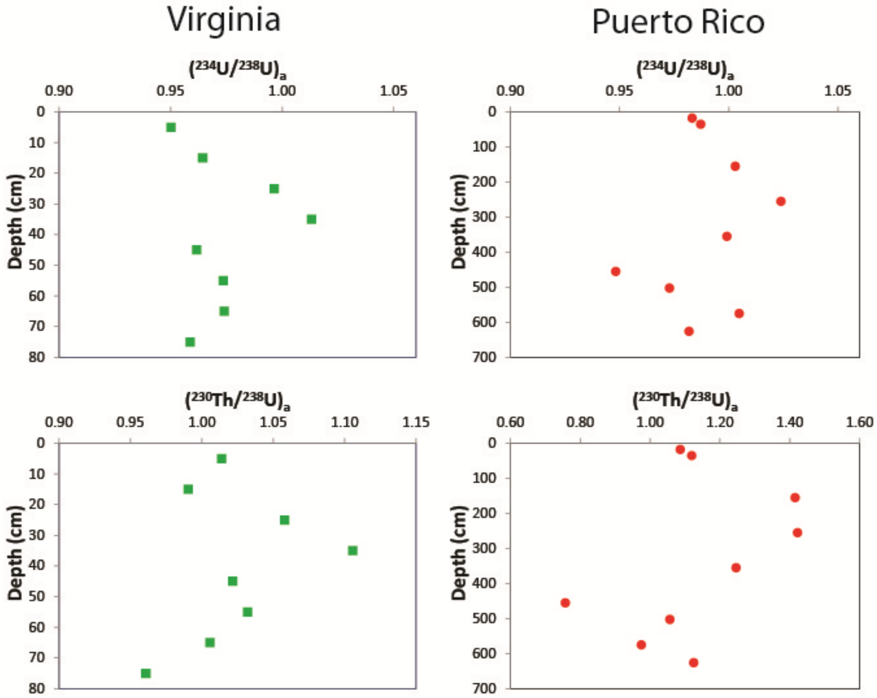

Fig.1 Preliminary results from parts of the Virginia and Puerto Rico profiles.

Furthermore, the ($^{234}U/^{238}U$) and ($^{230}Th/^{238}U$) values in the deeper Virginia profile increases from 80 cm to 40 cm depth, suggesting accumulation of both isotopes at 40 cm depth (Fig. 1). A more scattered fractionation pattern is observed from 600 to 300 cm depth in the Puerto Rico profile. One possible reason for the lack of clear trends here is that only 9 samples were analyzed from the total 600 cm thick Puerto Rico profile (Fig. 1). More sample analyses from the deep profile will greatly help to define the variation patterns in more details for this thick profile. It is also possible that U series disequilibrium is a technique that may not be useful in more tropical climates: ascertaining the answer to this question is one of the goals of the proposed work. An improved understanding of what causes these complex fractionation patterns in the Puerto Rico profile is definitely a great need for further applications of U-series disequilibrium to determine regolith formation rates. For example, changes of mineralogy, organic matter, and redox conditions in thick profiles might explain some of the fractionation patterns. Such factors can be examined by combining the new U-series isotope data set with the existing major and trace element and mineralogy datasets.

Furthermore, more sample analyses from ridge top sites in other catchments along the shale weathering transect will reveal whether we have simple or complex fractionation patterns of U-series disequilibrium along the climate transect. The comparison of all ridge top sites will help to understand why simple fractionation patterns of U-series isotopes are observed (such as at the Shale Hills catchment) vs. why complex patterns are discovered (e.g. at Virginia and Puerto Rico), and whether the simple or the complex fractionation patterns are the norm.

Hence, this seed grant will help to support the continuation of sample analyses from the existing sample archives at Penn State. Two undergraduate students from UTEP will be supported to analyze additional ~ 40 samples for U-series isotopes. The proposed work is expected to be complete by the end of summer 2013. PI Ma and UTEP undergraduate students will work closely with Penn State collaborators on data interpretation and publication.

Anticipated results and outcome

Measured U-series isotope compositions in these profiles will show disequilibrium values as expected from shale weathering processes. Combined with the existing major and trace element data and the observations of soil properties in these profiles, the behavior of U-series isotopes during chemical weathering and the controls of U-series disequilibrium signatures will be characterized. The degree of U-series isotope disequilibrium will be used in a U-series mass balance model similar to Ma et al. (2010; in review) to quantify the timescales of chemical weathering and calculate regolith formation rates, once the controls on U-series isotope fractionation are characterized and the reasons for the complex patterns are better understood.

As a general trend in the mean annual air temperatures of these sites, we expect to see an increase of regolith formation rates from north to south along the transect. We then will be able to characterize the effects of air temperature on shale weathering rates. Rates of shale weathering will also allow for quantification of element release rates in these soil profiles using the mass balance approach in White (2002). For example, the existing major and trace element profiles will allow for the release rates to be calculated for C, P, Pt and REE group elements.

Furthermore, we propose to take advantage of the existing CZO network across the nation to investigate the controls of lithology on soil development. More specifically, the proposed work will provide regolith production rates on shale bedrock in Puerto Rico, where formation rates on soil profiles developed on granodorite and meta-volcanic rocks are available from literature. Our study will thus allow for comparison of regolith formation on different lithology for ridge top sites under similar warm and humid conditions. Our results will contribute towards a better quantification of the important factors that control regolith formation and chemical weathering. Finally, our proposed work will further develop this novel isotopic dating tool for Earth surface processes by utilizing the existing CZO infrastructure and network resources.

Proposed SSHO Interactions

This proposed scientific work focuses on understanding the rates and mechanisms of regolith formation along a shale weathering transect, established as satellite sites of the SSHCZO. This work will evaluate the climate control on shale weathering and will be able to better predict the effects of future climate changes on rates of shale weathering and their impacts on global geochemical cycles. It thus fits directly into the ongoing activities of the SSHO group that emphasize on providing *a quantitative prediction of Critical Zone creation and structure, focusing on pathways and rates of water, solutes, and sediments*. The research work will help to address one of the key science questions at Shale Hills: *what are the rates and mechanisms of important hydrological, ecological, and geochemical processes?*

The research work will involve two undergraduates from UTEP, a research-intensive doctoral university with a Hispanic majority student population. Undergraduate students will be trained in the U-series analytical laboratory at UTEP and learn various analytical techniques for isotope research in Earth and Environmental sciences. Collaboration between UTEP and Penn State will attract participation of undergraduate students from historically underrepresented groups in environmental research. This project will offer a great opportunity to help them to study soil formation and climate change, expose them to cutting-edge critical zone sciences, and prepare them for future career opportunities in the Environmental and Earth Sciences. In addition, the summer field school offered by Shale Hills CZO provides an excellent opportunity to support general environmental education and bring public awareness of critical zone sustainability. I will encourage participation of undergraduate students from UTEP in this learning experience, through their involvement in this project. Impacts also include support of the career development of an early career faculty member (PI Ma) in running the U-series analytical laboratory at UTEP.

This project will utilize greatly the existing resources at Shale Hills CZO. We will request subsamples of parent rocks and soils archived by SSHO groups from the shale weathering transect and analyze U-series isotopes at UTEP. We will discuss the new U-series isotope data set, in conjunction with the existing major and trace element data from these sites. Physical samples will be permanently catalogued and archived in PI Ma's research lab at UTEP.

I expect at least one conference abstract and one peer-reviewed publication from this seed grant, with collaborators at Penn State as co-authors. We will follow strictly the EAR data policy (http://www.nsf.gov/geo/ear/EAR_data_policy_204.pdf) to publish our dataset, no later than two years from acquisition/analysis. Data and metadata collected for this project will ultimately be made available to the public through the Critical Zone database. I will also make the unpublished data, including geochemical data and images openly available, through creating a web-based data file on PI Ma' s research websites and the international Critical Zone Exploration Network (www.czen.org). Announcements of new publications and datasets will be made in a timely manner to inform the research community through the CZEN mail list.