**Intimate coupling among mechanical stresses, weathering and lithology control the fracture patterns within the Shale Hills Critical Zone Observatory**

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**ABSTRACT:**

Rock fractures are ubiquitous featureswithin the deep critical zoneand are expected to influencegroundwaterflow. The presence of rock fractures mediate interplay among pressure, temperature and chemistry. Despite substantial research, no general framework exists for systematically predicting the distribution of fractures within watershed settings, which may contribute to flow. In this paper, we explore the cardinality between fractures and fluid flow by coupling geophysical and geochemical analyseswithin the watershed of Susquehanna-Shale Hills Critical Zone Observatory (SSHO).Observed fracture patterns based on geophysical measurements and comparative analyses of major element concentration of borehole core samples suggest that the physical breakdown of rock is anegotiated impact of lithological variation, chemical weathering, and mechanical stresses generated by tectonic movement and topographical burden.Even if they do not cause new fractures, they could activate and cause displacement on old fractures, making the rocks easier to erode and increasing the permeability.Natural gamma ray logs are found consistent with the identified fracture patterns using Optical Televiewer (OTV), which show significant fracturing at a shallow depth of ~3m to 7m within the valley, and relatively less fracturing uphill on the ridge tops.It is expected that similar lithology should follow stratigraphy of the catchment where the bedrock is dipping towards north west. We note that fracture orientations likely control groundwater flow within the watershed and the aqueous pathways are the resultant of multivariate orthogonal function interconnecting spatially distributed heterogeneous fracture abundances.The hydraulic conductivity values implicate spatial variability within the watershed which decreases from valley floor to ridgetop.

**Keywords:** fracture orientation; lithology; weathering; natural stress; hydraulic conductivity; Shale Hills Critical Zone Observatory

**1. Introduction**

Among the most challenging and difficult problems in predicting flow are ones involving flow in fractured rock (Day-Lewis et al., 2003; Neuman, 2005). Significant research exists highlighting the importance of fracture flow in hydrologic response (Anderson et al., 1997, Montgomery et al., 1997, Ebel et al., 2008)and these features strongly affect the sediment erosion and rate of weathering (Hickman et al., 1995; Wibberley and Shimamoto, 2003).In large systems such as watersheds, it can be difficult to obtain sufficient geometric and hydraulic information in the field to constrain the connectivity of fracture networks for inclusion in modeling, which leads to bedrock being considered impermeable in many watershed-scale numerical models (Eaton and Bradbury, 2003; Kwon et al., 2004). Simulating groundwater flow and solute transport in these settings is therefore a marked challenge given the wide ranges of hydraulic conductivity values and relevant spatial scales that occur (Reddell andSunada,, 1970; Cooley et al., 1986; Hart et al., 2006).

In groundwater studies, the hydraulic properties of fracture sets are commonly represented using either a discrete fracture network (e.g., Sudicky and McLaren, 1992, Bogdanov et al., 2003a, Bogdanov et al., 2003b, Casciano et al., 2004, Cvetkovic et al., 2004, Helmke et al., 2005) or a continuum approach (e.g., Hsieh and Neuman, 1985, Wellman and Poeter, 2005; Wellman et al., 2009). The single most compelling reason for modeling fractures discretely is to explicitly capture the connectivity of flow paths in thesubsurface using a geologically realistic representation of heterogeneity (Eaton and Bradbury, 2003). Despite the benefits of such models, it is usually difficult to obtain measurements of the geometric and hydraulic properties of individual fractures in the field to calibrate these models in detail for predictive purposes (e.g., Berkowitz, 2002;Neuman, 2005). A driving factor may be where data are available: models of discrete fracture networks may make themost sense in systems where fractures are large and sparse, and fracture orientation and aperture can be characterized, but such characterization may be limited over the large spatial extent considered in most watershed models.

The geometry of fractures in the watershed is controlled by many factors. The most dominantfactors are: the subsequent changes in stress brought about by natural processes (e.g. tectonic activity, topographic stressand deformation of fractures owing to heating and cooling);mineral precipitation and dissolution as fluidflowsthrough the fractures; and lithology of the watershed (Roberts and Einstein, 1978; McTigue, and Mei, 1981; Savage et al, 1985; Pyrak Nolte et al., 1988; Olsson 1992; Miller, and Dunne, 1996; Molnar, 2004; Molnar et al., 2007). However, these processes are poorly characterized in terms of fracture development that governs the water movement across the complex and heterogeneous watershed. The Susquehanna-Shale Hills Critical Zone Observatory (SSHO)was established to investigate the creation, evolution, and function of regolith within a forested catchment lying on shale, including the movement of water throughout this system. It provides an excellent field laboratory to better conceptualize the fracture pattern and behavior, where the fractures are present within the deep critical zone. The dynamics of water within the system is needed to date groundwater, estimate soil-weathering rates, and quantify the residence time of solutes in the subsurface, among other processes (e.g., Amundson et al., 2007, Brantley et al., 2007, Anderson et al., 2008). Despite the understanding that characterizing the connectivity of hydraulically conductive pathways is critical for predicting the transport behavior of complex materials (e.g., Caine and Tomusiak, 2003; Knudby and Carrera, 2005), the controlsof the fractured bedrock on transport and mechanical weathering within the watershed has only been explored in a limited context (e.g. Brantley et al., 2011 and 2013; Jin et al., 2010; Kuntz et al., 2011; Jin and Brantley, 2011; Ma et al., 2011). Mechanical weathering (e.g. Lore et al., 2001) along with chemical weathering has been recognized to promote the development of regolith and have a positive feedback cycle with respect to enhancement of permeable pathways within the watershed (Carson and Kirby, 1972; Millot et al., 2002; Braun et al., 2005; Molner et al., 2007; Jin et al., 2011a, Jin et al., 2011b). Therefore geochemical alteration of shale provides an important hint on fracture flow within the watershed scale.It serves as chemical control which causes new fractures and enhances old fractures, in which dissolution may lead to widening and dilation may lead to irregular extension of cracks (Gabet et al. 2006, 2010; Royne et al., 2008; Jamtveit et al., 2011).

Superimposed on the existing fractures are stresses arising from neotectonics and topography that may influence flow. We hypothesize that topographic stresses influence catchment hydrology in a predictable way by triggering new fractures or reactivating and opening existing fractures. A few studies have investigated whether topographic stresses can influence groundwater flow. For example, Morin and Savage (2002) used an analytical model of Savage and Swolfs (1986) to estimate the stresses induced by ridge-valley topography surrounding two water wells in west Texas, one located in a valley, the other located on a ridge flank. They found that the transmissivities of lithologic units differed by up to a factor of ten between the wells, with a sign consistent with the predicted stresses. Morin et al. (2006) used a finite-element model to calculate stresses in an aquifer beneath a 220 m deep valley in Nova Scotia, and found evidence that topographic stresses open pre-existing fractures that are favorably oriented with respect to the induced principal stresses and close fractures with less favorable orientations. Martel (2000, 2006, 2011) emphasizes the potential for sites with topographically induced fractures to develop highly conductive flow paths with predictable orientations. However, these studies mostly focused on deep (>100 m) process rather than the more shallow critical zone.

It is important to study the lithological variation within the watershed to predict the rock fracture distribution. Lithology is a significant factor that controls the fracture intensity within the watershed scale. Shale is characterized by parallel layering that breaks competently along the bedding planes (Blatt and Robert, 1996). Whereas, sandstone and mudstones, on the other hand, are similar in composition but do not show the fissility. Therefore the influence of stress over shale formation is relatively higher than mudstone and sandstone in terms of fracture generation. This property of shale provides a favorable path for weathering reaction fronts that intensity during water-rock interaction.Fracture prediction is one of the most difficult studies that represent physical, chemical and hydrological coupling over time.

Fracture sets typically occur as groups of tens to thousands of individual fractures; however, only a small proportion of those may be relevant for conducting fluids (Long et al., 1991;Renshaw, 1995; Hsieh and Shapiro, 1996; Johnson, 1999). In this work, we explore influence of subsurface fracturing on flow by integrating geophysical and geochemical observations. Documented field observations (e.g. Fisher et al., 2008; Grob and Van der Baan, 2011) focused on changes in fracture distributions across lithological boundaries that allow us to identify the lithological variations across the watershed and its relation to fracture distribution. We look to address whether topographic stresses influence catchment hydrology by triggering new fractures or reactivating and opening in existing fractures. Coupling chemistry and geophysical data we try to characterize the geology and fracture distribution within the SSHO. We present the geologic setting within the SSHO and explore the mechanisms by which stresses, lithology and weathering may influence permeability within the watershed. Based on these mechanisms, we look to address the controls on flow within the SSHO.

1. **Methods**

**2.1Site Description**

The Susquehanna Shale Hills Observatory (Figure 1) is a 7.9-hectare forested site in the Valley and Ridge Physiographic Province of the central Appalachian Mountains in central Pennsylvania, USA. It is a first-order, V-shaped basin characterized by relatively steep slopes (25-35%) and narrow ridges.The ephemeral stream within the basin flows approximately west, and the major side slopes of the basin have almost true north-and south-facing aspects. Elevation ranges from 256 meters above sea level at the outlet to 310 meters at the highest ridge (Lin et al., 2006). The side slopes are periodically interrupted by seven un-channeled topographic depressions (swales). The stream is a tributary of Shavers Creek, which eventually flows into the Juniata River, a part of the Susquehanna River Basin.

**2.2 Geological observations**

The SSHO is mostly underlain by the Silurian Rose Hill Formation, locally consisting of ~250 meters of fossiliferous shale marked by well-developedfracture sets, and limestone and fine sandstone interbeds in the upper third of the formation; the dominantly shale formation is overlain by the thin (meters in thickness) Keefer (Sandstone) Formation toward the mouth of the drainage (Flueckinger, 1969). Some earlier studies indicate that the dip of the shale in the basin is quite steep (Fold, 1960, Lynch, 1976), while others indicate a more gentle dip (Hoskins, 1973), but these studies were based only on bedrock exposures outside of the SSHO catchment.

We present a third interpretation based on focused field work in and around the SSHO: steeply dipping strata with meter-to decimeter-scale folds superimposed. While each of these proposed settings is logical and consistent with the SSHO’s location within the valley and ridge physiographic province (e.g., Faill and Nickelsen, 1999), each would have a different effect on the development of subsurface fracture heterogeneity in the study basin.

The presence of gently dipping strata can be predicted given a simple trigonometric relationship between the known stratigraphic thickness of the RoseHill Formation in central Pennsylvania (ranging from 185 to 250 meters) and the horizontal width of the mapped subcrop belt (570 meters as determined from the Pine Grove Mills 7.5 minute quadrangle preliminary geologic map, where a subcrop is mapped bedrock beneath surficial deposits). This provides an estimate of dip ranging from 19º to 26º northwest, values that are congruent with the gentle dips suggested by Hoskins (1973). However, extensive field observations at eleven outcrop exposures in the unnamed tributary drainages to Shavers Creek,the creek adjacent to the SSHO, as well as from forty hand-dug pits to bedrock used for field soil descriptions in the SSHO, provide dip measurements ranging from 40 to 88º (average is 64º) northwest, indicative of much steeper bedrock dips in the study area than those inferred from the geologic map (Figure 2). When considered on a line of section approximately perpendicular to bedrock strike and parallel to the intermittent stream in the center of the SSHO watershed, those measurements are arrayed in an alternating steep (~70-88º) to less steep (~50-65º) series (Figure 2). This series is suggestive of the presence of meter-to decimeter-scale folds superimposed upon the overall steeply dipping strata, the preferred structural setting presented here. This conclusion is supported by the presence of meter-to decimeter-scale folds in outcrops of the Rose Hill Formation at nearby Greenwood Furnace State Park (Figure 3a-c), at Barree, PA, where steeply dipping Rose Hill Formation strata are overlain by folded (and in places, the gently dipping fold limb of) KeeferFormation sandstone beds (Figure 3d), and approximately one mile to the northeast of the SSHO along bedrock strike where a tight syncline of Rose Hill Formation shales and Keefer Formation sandstone exists in the same stratigraphic position as the SSHO boreholes.

In Figure 4, three fracture sets are displayed: bedding parallel (striking ENE), slightly oblique to bedding (striking ~ENE), and perpendicular to bedding and strike (striking WNW). The WNW-trending fracture set, or cross-strike joints, record the trajectory of the stress field at the time of propagation (Engelder and Geiser, 1980) that is, prior to Alleghenian folding. Bedding parallel and oblique to bedding fracture sets, i.e. the shale fissility, were formed as compression of the fine-scale depositional laminae was released during erosion-induced unloading. Bedding parallel fracture sets are the most commonly seen in this area.The regolith extends 1 to 3m below the land surface of the valley floor and thins toward the ridgetop.

**2.3 Drilling core**

We drilled 7 to 8 meters deep boreholes (CZMW5-7) into the bedrock at the ridge top of the catchment in August 2012 using a direct rotary air drill. The boreholes, CZMW5, 6 and 7 were drilled on the northern side, eastern side and southern side of the ridge top respectively. Two relatively deeper boreholes at the northern ridge of the catchment, DC0 (50m) and DC1 (25m), were drilled in 1970 (prior to the establishment of the SSHCZO) and in 2006 respectively. In 2006, four additional boreholes (CZMW1-4) were drilled in the valley floor with direct rotary air drill to the depth of 16 m. These four drilled wells are closely spaced (3>m apart). While CZMW1-3 are located on the northern side of the first order stream, CZMW4 is located on the southern side. Another borehole, Lynch well, was drilled in the valley floor in 1970 which is ~100m away from CZMW1-3. In April 2012, eight ~3 m deep boreholes were drilled in the catchment, four (GP 19-22) are in the north facing slope (NFS) and other four (GP 23-26) are in the south facing slope (SFS).

Majority of the boreholes (CZMW 1-4, DC0 and GP 19-26) are cased with 4 m PVC pipe. While CZMW 1-4 are PVC cased up to 3 m depth, Lynch well is metal cased at the top and DC0 is thoroughly metal cased. Figure 5 illustrates the locations of boreholes within the catchment and Table 1 describes the details of the boreholes along with various studies performed.**2.4 Hydraulic conductivity measurement**

Slug tests were performed to collect the hydraulic conductivity data in CWMZ1, 2, 3,4, GP26 and DC0where DC0 is located close to DC1(Table 2). Hvorslev slug test method (Hvorslev, 1951) was adopted to evaluate the hydraulic conductivity of the catchment.

(1)

where, K is hydraulic conductivity (m/s), r is the radius of well casing (m), R is radius of well screen (m), Le is the length of well screen (m) and Tois time takes for water level to fall 37% of initial change (s). **2.5 Borehole geophysics**

OTV and Gamma log were performedto characterizethe fractured pattern, lithologyand weathered zonesof the watershed. While OTV data were collected from CZMW1 – 7 and Lynch well, gamma data were collected from GP19-26, DC1 along with all the CZMW and Lynch boreholes. Table(1)describes the various geophysical methods and theboreholes details.

*2,5,1Gamma logs*

Naturalgamma loggeris a tool where a scintillation detector measures the total natural gamma-ray emission from the borehole walls, which is associated with uranium-238, thorium-232, and potassium-40 (Paillet and Crowder, 1996).The resolution of gamma ray log is 0.1m and detects physical properties of rock particularly in shale formation. It is assumed that the intensity of gamma radiation per unit mass of the radioactive minerals is constant, and the measured activity is the sum total of individual source (U-238, Th-232, and K-40) with fractional mass (M1, M2 and M3 respectively). The radioactivity is proportional to the weight concentration of the radioactive minerals:

(M1A1)/𝝆+(M2A2)/𝝆+(M3A3)/𝝆 (2)

where M is the unit volume mass of radioactive minerals of activity (A), and ρ is the bulk density of the rock.

*2.5.2 OTV logs*

The primary components of OTV logging tool are the white light source, mirror, magnetic compass and a photoelectric transformer. OTVisused to acquire a geometrically accurate and magneticallyoriented 360-degree photographic image of the borehole wall as reflected in a hyperboloid mirror.The logging speed was 1m/min, selected based on the vertical and horizontal resolution criterion. The intersection of a fractured plane with the borehole creates an ellipse; unrolling the ellipse in 2D produces an oriented sine wave where the amplitude of the sine wave is the dip angle of the fracture (Figure 6).The image is oriented, so the dip and strike of the fractures and bedding planes intersecting the boreholeswere evaluated.All CZMW boreholes and the Lynch Well were loggedto provide optical, true color images of borehole walls that enable detailed measurements of fractures.Fracture density, orientation and dipof the bedding planes intersecting the wells of boreholes werecalculated. WellCad software was used to process all the geophysical wire line logs.

**2.6 Geochemical analysis**

* + 1. *Major element analysis*

We collected drilled core samples from CZMW 5, 6 and 7 borehole sites and cuttings were bagged, labeled for appropriate sample depths, and air dried. Most of the CZMW5 samples were olive brown in color, whereas most of the CZMW 6 and 7 samples were olive gray in color. At the CZMW 5, 6 and 7 siteat the ridge top, the soil is very shallow (~0.1m) and all the samples deeper than 0.1m at the sites are shale bedrock. The entire bulk samples for each site consisting of both granular powder and rock fragments and was ground to pass through a 100-mesh sieve (150 μm).According to the method describe by Feldman, (1983), samples were prepared for lithium metaborate fusion to determine bulk chemicalanalysis by inductively coupled plasma atomic emissionspectroscopy (ICP-AES) for major elements at the Pennsylvania StateUniversity. These analyses for major elementsare estimated to have a precision of ±3% (Brantley et al., 2013). The method used in the present study was identical to the previous studies for CZMW1-4 and DC1 core samples (Jin et al., 2010, Jin et al., 2011a, Brantley et al., 2013).

According to Jin et al., (2010), DC1 samples were constant in element concentration and the standard deviation of the major elements concentration around the mean were observed to be relatively low. Based on the argument, the average DC1 composition observed by Jin et al., (2010) was used as the parent material for the present study. In this study, we assume Zrconcentration inthe bedrock is not removed or added throughout the profile, i.e., it is an immobile element. Zrwasmeasured with an error of 10 ppm with the ICP-AES technique.

* + 1. *XRD analysis*

X-ray diffraction technique is a powerful tool to identify the minerals in rocks and soils (Reynolds, 1989). XRD has long been a mainstay in the identification of clay minerals in soils (Jackson, 1956; Brown and Brindley, 1980; Whittig and Allardice, 1986). PANamalytical Empyrean series 2 was used to produce X-rays by the rapid deceleration of fast-moving electrons for XRD analysis that consist a X-ray tube with a filament electron source and a metal target. Activation of the tube entails passing a current through the filament establishing 40 mA current under 45 kV voltages between the filament and target that generate X rays.

We used mineral powder diffraction data to identify minerals composition of the catchment. Sample preparation protocol is described by Eberl et al. (2009).To establish structural details of the minerals present in the powdered samples,information of diffraction angles and relative peak intensities of the minerals were used. All sample preparation steps are intended to maximize random orientation to increase the exposed surface area of the included minerals that generate multiple diffraction peaks. Mineral phases were identified with JADE software.

**4. Results**

**4.1 Wireline logging** *4.1.1 Ridge top*

The OTV data for the boreholes illustrate existence of partial fractures and minor open fractures in the ridge top boreholes (CZMW5, 6 and 7). No major fractures were observed upto a depth of ~7m below the surface. This is supported by the gamma data which exhibit a relatively lower standard deviation in gamma count varying 11 to 14 counts per second. Gamma log of DC1 which is comparatively deep (~23m) the borehole show a lower rate of average gamma count and higher standard deviation than CZMW 5,6, and 7 (Table 3). Jin et al. (2010) reported that the density of shale chips recovered from DC1 was much more variable above 5-6m depth than those below and also abrupt increase (compared to deeper depths) in porosity was measured via neutron scattering in drilling fragments at 5 m depth. Jin et al. (2011b) described varying dissolution fronts down to approximately 25 m; it may also indicate a change within the Rose Hill Shale and may be indicative of similar processes underlying the ridge top.

OTV data of CZMW 5, 6 and 7 reveal that the fracture dips are in the North West direction and dominantly follow the bedding pattern of the watershed (~ 40o to 55o). Figure 7 (a, b and c) reports the fracture orientation, bedding dip and rate of gamma count of boreholes CZMW 5, 6 and 7 respectively.Only gamma logging was performed in DC1 and shown in Figure 7(d).

*4.1.2 Slope*

The average gamma count decreases towards the valley for the boreholeslocated in the slope. Figure 7(d) illustrates while the mean gamma count gradually decreases from ~155 to ~139 in the south facing slope (SFS), itdecreases from~159 to ~124 in the north facing slope (NFS).Topographic profile of the slope reveals, SFS has an average gradient of ~150 whereas the NFS exhibits a gradientaveraging ~200 (Figure 5). In ideal case, chemical weathering is driven by hydrological flux and weathering is inversely related to the slope gradient. This possibly suggest the relatively steeper NFS to be less weathered and can be explained with the higher values of observed average gamma count per second (Table 3).

*4.1.3 Valley floor*

CZMW1, 2, 3, 4 and Lynch well are located at the valley floor and while ~ 3m is pvc cased for CZMW1- 4, Lynch is iron cased until ~2.5 m below the surface.The OTV data demonstrates higher abundance of fracture between ~3 to ~7m of depth below the surface and is supported by gamma data. This also explains why the R2 values for the boreholes (CZMW1-4) at the valley floor hold a very good correlation (> 0.8) with the depth (Figure 8) unlike other CZMW5, 6 and 7 boreholes which are located at the ridgetop.The majority of the fracture dips of CZMW 1, 2 and Lynch well are parallel to the bedding plane in the NW direction as shown in Fugure 7(e, f, and i). However the fracture orientation of CZMW3 and 4 exhibits a random pattern as shown in Figure 7(g and h).OTV logs of the borehole indicate the bedding dip varies between 40 and 50 degrees.

Kuntz et al. (2011) note in the CZMW wells in the valley floor that the variability in natural gamma increases near the surface, and attribute this phenomenon to the weathering out of clay materials within the highly fractured and broken part of the subsurface from a depth of ~7 m to the surface; the gamma is therefore assumed to be a proxy for mass loss (minus compaction). Although variability in gamma count may also be a function of sandstone interbeds seen in the optical televiewer logs (Figure 7 e-i).

Drilling and well log data reveal a calcareous slow drilling zone around 6-7 m below the land surface (Ma et al., 2011) above which is highly weathered saprock with comparatively high fracture density, and beneath which is a less-fractured, and rather geochemicallyhomogeneous blue-grey shale. Below 7 m, the televiewer indicates that the coloring of the shale is more consistent, and the well is less fractured beneath this point. Both the gamma logs and OTV logs are well correlated to the fracture occurrence within the valley.

However we find a relatively moderate facture profiles atthe relativelyolder Lynchborehole in the valley,roughly 100 m upstream of the CZMW 1-4boreholeswithin the watershed. We see a continuously distinct decrease in gamma-ray emissions(less than ~20cps) than that seen in the other CZMW1-4 wells (Table 3) in the valley. The gamma logs qualitatively describe the presence of sand interbed and missing clay within the system. This also suggests a possible instance of weathering due to the exposure for an extendedperiod of time.

Figure 9 shows the depth vs gamma ray count rate of all the wells together thatrevealsthe GRCR varies between 140 and 195 at the ridge top and gradually decreases towards valley. The GRCR is fluctuating between 123 and 175 in the south facing slope and between 115 and 180 in the north facing slope. Thesedata support the OTV logs that indicate the 1D zone (ridge top) is relatively less fractured. Whereas, 3D zone (valley floor), is characterized by a higher degree of fracturing. Contrary to the 1D zone (ridge top), the frequency of sealed fracture are less and most of the upper parts in 3D zone are characterized by higher degree of fractures.Fluid flow in the regolith is considered to be largely two-dimensional downslope, i.e., non-convergent flow, and the hill slope is thus can be referred as a 2-D weathering profile which shows relatively lesser fractured/weathered zone.

Lower hemisphere stereographic projection of fracture dip (Figure 10) indicates that the majority of the fractures follow the NW direction along the bedding plane, except for CZMW3 and CZMW4 where fracture orientations are comparatively random. The fracture systems are greatly influenced by geological origin of the rocks. For example, tectonic force and topographical burden drive the normal and shear stresses across the watershed which breaks the shale formation along the bedding plane and presence of water making the rocks easier to weather and erode and increasing the permeability. This advocates that the presence of fracture abundances and water allow the physical stresses and chemical weathering to break the weak rock in random directions.

**4.2 Geochemical analysis**

*4.2.1 Major element analysis*

Chemical weathering of the bedrock mobilizes elements and leaves important clues to understand the underlying geochemical behavior. We calculated the mass transfer coefficient (𝜏i,j) to evaluate the element addition or depletion from the bedrock.

(3)

Where C is the concentration of immobile element (i) and mobile element (j), relative gain or loss of mobile element was evaluated with comparison to immobile element. Positive mass transfer coefficient values reflect the addition of element, negative values define depletion of elements and zero indicate the immobile property of the element with respect to parent material.

Al, K, Mg and Fe are soluble major elements that commonly associated with primary clay minerals, e.g. illite and chlorite (Jin et al., 2010).The observed depletion profile of the major elements selected from the core samples of CZMW 1-7 and DC1 (Figure 11) depict that depletion of soluble major elements in comparison with the parent material in the valley is significantly higher, than that in the ridge top. In other words, clay mineral dissolution is identified higher in the valley from surface to 7m depth which is associated to the densely fractured area indicating geochemical alteration of clay under aqueous conditions and fractures influencing the weathering processes. This also suggests a possible vice versa relation between fluid flow and fractures where fracture encourages fluid flow which in turn provide a positive feedback channel to influence fractures. Even if weathering does not cause new fractures, they could activate and cause displacement on old fractures, making the rocks easier to erode and increasing the permeability.

*4.2.2 XRD analysis*

Qualitative X-ray diffraction mineralogical data of core samples selected from CZMW2, CZMW5, CZMW6, CZMW7 and DC1 (at the depth of ~7m below ground surface) show that Rose Hill shale is composed predominantly of Quartz, Illite, Chloride, Calcite, Plagioclase and K-feldspar. CZMW 2 sample also contains Hametite, Ankerite (Fe and Mn rich carbonates) and Pyrite. Hametite is also identified in CZMW5, CZMW6 and CZMW7 that is associated with gray color of the formation. The results exhibit higher abundances of clay minerals in all the samples (Table 4).

**4.3 Hydraulic conductivity**

The hydraulic conductivity of a core of the shale bedrock matrix is very low; Kuntz et al. (2011) sampled Rose Hill Formation shale from an outcrop ~15 km NE of the SSHO and found a value of ~10-15 m/s using the transient pulse-decay method (e.g., Brace et al., 1968; Lin et al., 2006) in a tri-axial pressure apparatus. The open valley wells indicate an in situ hydraulic conductivity of ~10-6 m/s, averaged over the uncased portion of the wells between 3 and 16 m depth. It gradually decreases from the valley floor towards the ridge top. Slug test results indicate that the GP26 exhibits a hydraulic conductivity of ~10-7 m/s, whereas it is~10-9 m/s for DC0 (Table 2).This result indicates that fractures dominate the measured in situ permeability of the site. Slug testing of DC0 indicates that the hydraulic conductivity at the open part at the bottom of the well is approximately two orders of magnitude less permeable than in the valley, but we note that this measurement of hydraulic conductivity is only valid for the small depth of open well at 50 m

**5. Discussion**

The rocks in the SSHO are heavily folded and fractured, and our observations suggest that fractures are dominant controls on groundwater flow in bedrock in this setting. Fractures are a function of three major factors: lithological structure and the tectonic environment of formation, present-day stresses arising from neotectonics and topography, and weathering. We observe steeply dipping strata (~40o-55o) in televiewer logs and steeply dipping strata based on measurements made around the basin and from soil pits, and suggest that these observations can be reconciled with a structural setting consisting of steeply dipping strata with meter-to decimeter-scale folds superimposed, similar to structures observed in nearby outcrops. It is expected that fractures normal to bedding are formed during folding and bedding parallel features are formed due to topographical stress and weathering process. The bedding parallel fissility likely gets accentuated as weathering occurs near the surface, preferably in valley floor where hydraulic head is near the surface and the formation of clay minerals is involved. Whereas deep fractures may be the result of near-surface topographic stresses, where weathering is likely lower than at the surface.

Figure 12 is the conceptual two dimensional representation of the catchment that summarizes the comprehensive understanding of the fracture pattern within the catchment. The observatory is developed almost entirely upon Rose Hill (Folk, 1960; Lynch, 1976), a thick Silurian-age formation (Lynch, 1976) where regolith on the ridge top is relatively thin (~0.3m) and extends to several meters (>3m) across the valley floor. Hydraulic head is shallow (near the surface) in the valley and deep in the ridge top (Brantley et al, 2013). Bedrocks are dipping (~40o-55o) towards the North West direction and cut obliquely across the catchment that is parallel to the intersection of sand stone keefer and rose hill shale (Figure1). It was observed by OTV that the fractures pattern dominantly follow the bedding plane whereas fracture density is function of topographic stresses and lithological variation, due to the presence of shale, mudstone, and sandstone layers. The effects of topographic stresses are superimposed on the mechanisms of lithology and weathering that influence rock fracture. There may be a decline in fracture abundance with depth throughout the landscape, whereas topographic stresses modulating this trend, resulting in and gradual decline in fracture density beneath the valleys and gradual increase beneath the ridge tops. The prevalence of fracturing within the bedrock likely serves as an important control on the “active depth of flow,” considered to be the zone in the subsurface that responds to annual recharge and climatic variability and has groundwater residence times that become older from the recharge to discharge areas (Mayo et al., 2003).

In the gently dipping features near the outlet of the watershed, infiltrating groundwater would encounter fractures sub-parallel to the nearly horizontal bedding, so the component directing water downward would be smaller than elsewhere in the watershed where more steeply dipping features are expected. Gentle dips appear only in the upper part of the stratigraphic section where the Keefer Sandstone lies. Down stratigraphic section (on the ridgetops and farther upstream within most of the watershed), dominant fracture sets are steep and parallel or slightly oblique to bedding, and therefore may provide a better pathway for water infiltrating into the subsurface. These steeply oriented fractures provide flow paths down slope to the SSHO stream, but are perpendicular to the flow gradient toward Shavers Creek (Figure 1). In the upper part of the stratigraphic column, in the valley, fractures are mostly bedding-parallel and gently dipping, and dip down stream, thus providing a flowpath toward Shavers Creek. This change in the dip of strata, suggested to be a consequence of folding, likely produces anisotropy in the flow within the watershed.

Systematic differences in permeability were found beneath ridgelines and valleys that follow a general trend in elevated potential for rock fracture beneath valley floors in comparison with the ridge top. Higher fracture density in the depth beneath the valley floor that was observed in the OTV logs of CZMW1-4 boreholes that demonstrate the higher magnitude of hydraulic conductivity compared to the ridge top. Fractures are dominant within the depth of 0-7m below the valley floor and higher open fractures are present at the depth of ~5m in CZMW1-4 which possibly indicate the connectivity of the fractures. Valleys may act as “drains” for shallow groundwater flow through watersheds, potentially driving a positive feedback in which water penetrates deeper and faster into deeper valleys, accelerates the weathering that breakdown the bedrock, and enhances the growth of valley relief.

Borehole gamma-ray emission measurements demonstrate the feasibility of a weathering feedback driven by fracture abundance. The roles of landscape positions and hydrological processes on fractured rock during weathering are assessed by comparing weathering profiles among three different profiles (e.g. slope aspects, top of the ridge and valley floor within the catchment). In the valley, we show that a variable signal in gamma-ray emissions correlates strongly with fracturing, and is likely evidence of clay weathering out of the system. Natural gamma ray is directly associated with the sediment flux and represents the quantitative variability of the resident radioactive materials (isotopes of Potassium (K), Uranium (U) and Thorium (Th)). Fall in gamma ray count rates are associated with radioactive mass loss and weathering potential.

The shale bedrock is composed predominantly of illite (58 wt. %) and feldspar (plagioclase and Kfeldspar) which are K bearing minerals (Jin et al., 2010; Ma et al., 2010) and contribute toward the gamma count. U is soluble in water while Th is “particle-reactive”, i.e., it generally associates with solid surfaces (Langmuir, 1978; Langmuir and Herman, 1980). The weathering behavior of K–bearing minerals determines the radioelements of weathered rocks and soils. During weathering, major K hosts are destroyed and released K are taken up in clays which eventually flush out during weathering cycle. A relatively lower K content in a weathered zone attributes reduced radioactive responses towards the gamma ray logger and results in a relatively lower gamma ray count. An inverse relation is found to exist between GRCR and the increasing distance from the ridge top towards the valley (Figure 9). A possible explanation could be the lesser fractures abundance as we move upward from the valley floor towards the ridge top up to the depth of 7m from the surface. We observe that the gamma ray count of north facing slope is more than the relatively flat south facing slope which indicate higher rate of weathering in relatively flat south facing slope (~15o) than steeper north facing slope (~20o). This is in conjunction with the result observed by Ma et al., (2010), which calculates regolith production rates using uranium series isotopes.

Subsurface water flow through fractures promotes mechanical (e.g. expansion and contraction caused by freezing and thawing, heating and cooling, wetting and drying) and chemical weathering in direct way. The mass transfer coefficient analysis of major element in comparison of parent rock composition and their comparative results in terms of the valley and the ridge top site suggest the water front propagates into the bedrock through fractured bedrock cause progressive loss of clay minerals represented by major cations that accelerate the regolith formation rate.

**6. Conclusions**

Our results suggest that fracture orientations likely control groundwater flow within the deeper watershed. Topographic stresses, lithology and weathering are dominant controls on near surface fracture generation in present days and create spatially variable hydraulic conductivity within the critical zone. The geologic setting relevant to water flow is steeply dipping strata with meter to decimeter scale folds superimposed throughout most of the watershed, with more gently dipping features near the contact with the Keefer Sandstone. Patterns in the observational data provide evidence of highly variable fracture orientation and rock structure, and consequently imply a significant variability in the direction of flow.

Groundwater flow—in this case meaning flow beneath the water table and not considering the impacts of interflow in the vadose zone after precipitation—in the basin is likely to be anisotropic because of the combined effects of superimposed folds on the overall steeply dipping strata. In the majority of the watershed, the bedding parallel fractures are steeply inclined (with the steeply dipping beds) and oriented approximately perpendicular to the ephemeral stream in the SSHO watershed. Thus, while these fractures provide subsurface flow paths down slope to the SSHO stream, the orientation and steep dip do not facilitate fluid flow down gradient toward the trunk stream, Shavers Creek. These fractures help move flow toward the SSHO channel but not out the watershed. In contrast, near the outlet of the watershed the bedding parallel fractures are gently dipping downstream, so provide a subsurface flow path down gradient toward Shavers Creek, but may not act as important conduits of vertical fluid flow into the subsurface.

Topographic loading and unloading, mechanical weathering and progressive reactive fronts during chemical weathering process within the watershed help in connecting the distributed array of fractures and define the aqueous trajectories. We are unable to yet distinguish whether the natural stresses, weathering or lithological variation is more dominant factor to control the fracture occurrence that characterize the spatially variable hydraulic property within the watershed, especially at shallow depths beneath the ridge tops and the valleys. Field observations of variable fracture orientation and abundance, combined with other proxies, indicate that fracture orientations can vary enough within a watershed to significantly affect patterns of subsurface flow. Overall our study suggests that there is coupled interplay of these processes which is responsible for spatially and depth dependent variability in fracture density at the SSHO.

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**Figure captions.**

Figure 1. (a) Shaded relief map generated from a 1-m resolution LiDAR (Light Detection And Ranging) survey of the SSHO site. The blue line (ABC) indicates the location of the intersection between Keefer Sandstone and rose hill shale .Laser altimetry data were acquired and processed by the National Center for Airborne Laser Mapping (NCALM).

Figure 2. Bedrock geologic map of greater SSHO region with structural information measured from outcrop and soil pits. The black transect indicates the location of the cross-sectional profile of dips projected to the line using strike and dip information from field data. Steeply dipping strata with superimposed folds can be interpreted from these measurements.

Figure 3. (a-c) Outcrop of shales of the Rose Hill Formation at Greenwood Furnace State Park, PA, displaying meter-to decimeter-scale folds in the shales (highlighted with dashed lines). The photographs are oriented as an up-section progression beginning from the top right in a counterclockwise direction. (d) Outcrop of shale and sandstone of the uppermost Rose Hill Formation and overlying Keefer Formation at Barree, PA. Note the gently dipping southeast limb of the folds and steeply dipping northwest limbs. To the right (out of the photograph) only steeply dipping shale beds of the Rose Hill Formation are encountered.

Figure 4. Rose diagram displaying fracture orientations measured at various outcrops adjacent to the SSHO. The small arrow on the perimeter of the northeast quadrant marks the average strike of bedrock measured at these outcrops. The dominant fracture set is bedding parallel with a secondary set slightly oblique to bedding. Bedding normal fractures are observed but are much fewer in total number in the shales.

Figure 5. Borehole locations and topographic profile in the Shale Hills Observatory where elevation of CZMW1 is 260.5m, CZMW2 is 260.34m, CZMW3 is 259.8m, CZMW4 is 259.98m, CZMW5 is 302.3m, CZMW6 is 309.8m, CZMW7 is 299.76m and DC1 is 302.97m. Topographic profile along north and south slope shows that the maximum elevation of GP boreholes in south facing slope is 278.6m and north facing slope is 278.9m

Figure 6.Illustrates the principles of Optical Televiewer operation. Schematic illustration of a borehole a layer dipping south: a) 3D, b) 2D; and c) Illustration of the equivalent sinusoid and dip calculation

Figure 7. Optical televiewer image logs with fractures projection log, gamma log, structural log representing tad-poles, and polar projection of fracture and bedding dips from the CZMW boreholes and Lynch well in the valley and the ridge top. Rock in the valley floor is significantly fractured up to 7m, comparatively very less fractured in the ridge top. The strike and dip were measured from the structural logs of all wells. In these logs, features dip ~ 40-55 degrees and dipping towards N-W direction. Gamma logs show the natural gamma ray emission from the borehole walls and decrease in gamma count is either as a function of sandstone interbeds or weathering out of clay materials 7(a) CZMW5, (b) CZMW6, (c) CZMW7 (d) represents Gamma ray emissions from DC1at the ridge top, and GP boreholes located at the north and south transects (e) CZMW1 (f) CZMW2 (g) CZMW3 (h) CZMW4 (i) Lynch well

Figure 8. Depth profile of the fracture density for CZMW1-7 boreholes and Lynch well. It delineates a decreasing trend of fracture distribution with depth for CZMW1-4 . However no particular trend is observed for Lynch well and CZMW5-7 boreholes. There is significantly high fracture density in the valley floor whereas boreholes in the ridge top show lower fracture up to the depth of 7m. It is expected that the lithological variation (mudstone interbed) may be responsible for the lower fracture density in Lynch well.

Figure 9 (a) Mean gamma ray emissions from CZMW 1-4, GP19-26 and CZMW5-7 up to 3m depth (b) Mean gamma ray emissions from CZMW 1-4 and CZMW5-7 up to 7m depth. Note that in all cases, a decrease in gamma counts are noted in the valley floor and it increases from slope to ridge top, as a function of weathering out of clay materials

Figure 10 Polar diagram illustrating fracture orientations measured from OTV logs of CZMW boreholes and Lynch well in the SSHO. Most of the fractures planes are dipping towards north- west direction and average dip of the fracture plane is parallel to dip of the bedding plane (~40o-55o). Fractures normal to the bedding plane are also observed but are significantly fewer in total number. Fracture orientation of CZMW 3 and CZMW4 are randomly distributed in compare to other boreholes in the SSHO

Figure 11 Depth profile of mass transfer coefficient values of major elements (Al, Fe, K, Mg) for the ridge top boreholes (CZMW5-7 and DC1) and the valley floor boreholes (CZMW 1-4). 𝜏 (mass transfer coefficient) values for CZMW 1-4 indicate higher mass loss of major elements, which represent the clay minerals depletion, than CZMW 5-7 and DC1 up to the depth of 7m.

Figure 12 conceptual two dimensional representation of the catchment in X-Z plane which summarizes the comprehensive understanding of the fracture pattern within the catchment

Table captions

Table 1 Basic description of the boreholes and corresponding measurement techniques

Table 2 Hydraulic conductivity of the boreholes in the SSHO

Table 3 Statistical analysis of natural gamma ray count observed at the CZMW1-7, DC1 and Lynch well

Table 4 Identified minerals in CZMW and DC1 boreholes core at the depth of ~7m

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