



Particle fluxes in groundwater change subsurface shale rock chemistry over geologic time



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ABSTRACT

Most models of landscape evolution posit that particles leave the land surface by physical weathering (i.e., erosion) and solutes leave the subsurface by chemical weathering. They assume that erosion does not affect the rock and soil chemistry. However, in this study of a shale catchment we discovered that particles are mobilized out of soil and weathering rock and are transported through the subsurface, resulting in changes in the rock and soil chemistry. We studied the solute and particle fluxes during six storms in the Susquehanna Shale Hills Critical Zone Observatory in Pennsylvania, USA and compared those to the record in regolith chemistry. The stream's suspended particles primarily consisted of platy-shaped, μm -sized illite, commonly coated with patchy, amorphous, submicron-sized Al-, Fe- and Si-rich oxides. The chemistry of the stream particles always differed from that of surface soils except during intense dry-season rainstorms. Stream particles were chemically similar to the laboratory-extracted soil colloids at high discharge but to groundwater particles at low discharge, implying a central role of flow path variations in controlling subsurface particle transport. Zr was effectively immobile in Shale Hills. Regolith chemistry revealed that the cumulative effects of particle loss to depths of 5–8 m in the fractured bedrock zone were estimated to account for 58% of K and 24% of Mg losses. In shale landscapes, we propose that subsurface particle transport must be considered in landscape evolution models as an important contributor to changes in rock and soil chemistry over geologic time periods.

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1. Introduction

Losses of solutes and particles from the critical zone shape its architecture. Most conceptual and quantitative models of critical zone evolution describe loss of solutes throughout the weathering profile by chemical weathering and loss of particles from the surface by physical erosion (Anderson et al., 2007; Dixon et al., 2009; Riebe and Granger, 2013; Yoo and Mudd, 2008). However, many lines of evidence show that particles are also lost from the subsurface in some locations and that this loss depends on physical, chemical, and hydrological conditions (DeNovio et al., 2004; McCarthy and McKay, 2004; Ryan and Elimelech, 1996). Here, “particle transport through the subsurface” is used to refer to the suspension and movement of particles of any size through soil and bedrock: we do not refer here to episodic mass movement such as landslides or other such processes.

In most weathering models physical erosion has been assumed to not affect soil chemistry. Given that assumption, chemically immobile elements such as Zr, Ti, and Hf or relatively insoluble minerals have been used to quantify elemental losses by chemical weathering (Anderson et al., 2002; Brimhall and Dietrich, 1987). For example, the non-dimensional mass transfer coefficient ($\tau_{j,i}$) is now commonly used to quantify the loss or accumulation of a mobile element (j) by comparing its concentrations in parent and weathered materials normalized by the immobile element (i) (Anderson et al., 2002; eqn. (1)):

$$\tau_{j,i} = \frac{C_{j,w}C_{i,p}}{C_{j,p}C_{i,w}} - 1 \quad (1)$$

here C is a concentration of an immobile element (i) or a mobile element (j) of parent (p) or weathered material (w). The τ values vary from -1 (complete depletion) to 0 (no change) to positive values (accumulation). Riebe et al. (2003) further proposed that by combining the concentration of immobile elements and estimates of the total denudation rate (D) from cosmogenic nuclide meth-

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ods (e.g., ^{10}Be), the relative importance of chemical and physical weathering fluxes can be quantified (eqn. (2)):

$$W = D \left(1 - \frac{[\text{Zr}]_p}{[\text{Zr}]_w} \right) \quad (2)$$

here W , D , $[\text{Zr}]_p$, and $[\text{Zr}]_w$ refer to the chemical weathering rate, total denudation rate (chemical plus physical losses), and the Zr concentrations of parent and weathered materials, respectively. Furthermore, the authors define $(1 - \frac{[\text{Zr}]_p}{[\text{Zr}]_w})$ as the “chemical depletion fraction (CDF)” (Dixon et al., 2009; Riebe et al., 2004, 2003).

Physical erosion is assumed to mobilize particles that have the same chemistry as the originating soil. However, particles that are enriched in Al, Fe, Si, and sometimes Ti are observed to be redistributed from one horizon to another within soil profiles and have also been documented to be lost from catchments (Aguirre et al., 2017; Bern et al., 2011; Jin et al., 2010; Kaup and Carter, 1987; Taboada et al., 2006; Trostle et al., 2016; Yesavage et al., 2012). Such particle losses, furthermore, may be significant enough to alter Al and Fe concentrations in soils (Jin et al., 2010; Yesavage et al., 2012). For instance, Jin et al. (2010) investigated the chemistry of soil, bedrock, and stream water at the Shale Hills catchment in the Susquehanna Shale Hills Critical Zone Observatory (CZO) in Pennsylvania. They reported that Al and Fe of the soil and fractured rock were depleted with respect to the parent material, but the solute concentrations of these elements in the stream (0.45 μm filtered) were negligible. They hypothesized that these elements were likely mobilized as particles. At the same site, Yesavage et al. (2012) also analyzed the chemistry of soil pore water, groundwater, macro-pore water (i.e., preferential flows through the soil), and stream waters. They found that Al and Fe concentrations of the soil-pore waters showed no differences between unfiltered and filtered samples (0.45 μm) while those of the other sampled waters were much higher in the unfiltered samples than in the filtered samples. The authors, therefore, postulated that the particles transporting these elements were larger than the pore-sizes of the lysimeter cups (1.3 μm).

Recently, different approaches have been developed to quantify elemental losses via solutes and particles (Bern et al., 2015; Hasenmueller et al., 2017; Sullivan et al., 2016). For instance, Hasenmueller et al. (2017) proposed using Al as the immobile element to quantify the losses of the solute and particle fractions of a mobile element (j), $\tau_{j,\text{Al}}$. The authors argued that Al was mobilized mainly as particles; therefore, the elemental loss as solute was revealed by using Al as the immobile element. They then assumed that Zr was immobile and argued that $\tau_{j,\text{Zr}}$ estimated the total loss of the mobile element. The difference between $\tau_{j,\text{Zr}}$ and $\tau_{j,\text{Al}}$ becomes the fraction of elemental loss as particles (P_j):

$$P_j = \tau_{j,\text{Zr}} - \tau_{j,\text{Al}} \quad (3)$$

The authors employed this method to quantify the elemental loss as particles in the Missed Grouse catchment, a catchment that lies next to Shale Hills, in the Shale Hills CZO: more than half of the potassium (up to 75%) and magnesium (up to 63%) fluxes were attributed to particle loss.

In another treatment, Bern et al. (2015) proposed a dual-phase mass balance model. They used a ratio of two elements such as Ti and Zr that primarily move via colloids (i.e., water dispersible colloids; WDCs) but that display different affinity to the colloids. The authors adopted a definition of colloid as particles smaller than 1 μm (Sposito, 2006). They operationally defined these colloids (material between 3 kDa and 1 μm) by extracting them from soil samples by shaking soil samples in deionized water for 10 min in the laboratory. They assumed that, as weathering progresses, Ti and Zr concentrations in parent bedrock, weathered material, and

WDCs develop characteristic Ti/Zr ratios. Then, the authors quantified the elemental loss and gain as colloids via the mass balance approach using the Ti/Zr ratios of these materials. Bern and Yesavage (2018) used this model to study colloidal loss in Shale Hills. They extracted and analyzed WDCs from the Shale Hills soils and estimated that more than 90% of the total mass loss was via WDCs in the catchment. They also concluded that Zr was mobile and that the loss of WDCs resulted in Zr depletion in the soil by 12–51% with respect to the parent rock.

However, these previous treatments of subsurface particle losses and weathering have been indirect and only limited to the soil layer. The importance of particle transport in the evolution of the entire critical zone structure, particularly the underlying fractured/weathered rock zone, has not been previously assessed. In addition, in Shale Hills particles that are larger than a micron—and therefore do not qualify as colloids—may play a key role in Al and Fe losses (Jin et al., 2010). Here, we return to Shale Hills to explore the importance of losses of particles in the context of critical zone evolution. The primary objectives are to identify mechanisms of mobilization and transport of particles of all size, and to quantify the importance of subsurface particle loss at the catchment scale in both short- and long-term time scales. We accomplished these goals by direct observations of mobile particles in groundwater and in the stream at various hydrologic conditions and by comparing the mobile particle chemistry to the regolith chemistry.

2. Methods

2.1. Study site

The Shale Hills catchment (drainage area: 0.08 km^2) lies in central Pennsylvania (U.S.A.) and is underlain by Rose Hill Shale. Illite was the most dominant mineral of the parent bedrock (DC1) followed by quartz, chlorite, and trace amounts of feldspar and Fe-oxides (Jin et al., 2010). In the deepest layer (>37 m below the land surface under the northern ridge) in the parent bedrock, ankerite was found while near the stream outlet, the carbonate-rich layer lies at 6–8 m deep (Brantley et al., 2013). The mean annual precipitation is 1090 mm and the erosion rate estimated from meteoric ^{10}Be measurements of stream sediment is 15 mMyr^{-1} (Jin et al., 2010). The stream is ephemeral and discharge is measured at a weir located near the outlet of the catchment at a 10-min interval. Stream and groundwater chemistry observations have demonstrated that the contribution of regional groundwater to the stream discharge may be small and may become important only at low flow (Sullivan et al., 2016). Instead, most stream discharge is fed by water that has quickly flowed through the upper layers of disaggregated soil (~1.4 m thick) and through a highly fractured bedrock zone that extends to 5 or 8 m depth throughout the catchment (Fig. 1; Graham and Lin, 2011; Jin et al., 2011a; Lin et al., 2006; Sullivan et al., 2016). Jin et al. (2011b) proposed that the fractures of this layer may have developed primarily by freeze–thaw processes since the Last Glacial Maximum. Because of these porosity contrasts, the perched groundwater table (i.e., interflow) transiently develops at the interface between the highly fractured rock and underlying less fractured zone (Sullivan et al., 2016). At high flow, the dominant flow path is preferential flow via soil macro-pores and soil horizon interfaces in the unsaturated zone (Graham and Lin, 2011; Jin et al., 2011a; Lin et al., 2006) and interflow. During highly intense rainstorms or at the beginning of snow-melt, overland flow occurs (Lin et al., 2006). During the dry season, the stream dries out.

2.2. Sampling

We collected water and suspended particle samples at 0.5–48 h intervals during 6 storm events using three ISCO samplers in the

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