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A coupled field study of subsurface fracture flow and colloid transport

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SUMMARY

Field studies of subsurface transport of colloids, which may act as carriers of contaminants, are still rare. This is particularly true for heterogeneous and fractured matrices. To address this knowledge gap, a 30-m long monitoring trench was constructed at the lower end of sloping farmland in central Sichuan, southwest China. During the summer of 2013, high resolution dynamic and temporal fracture flow discharging from the interface between fractured mudrock and impermeable sandstone was obtained at intervals of 5 min (for fast rising stages), 30–60 min (for slow falling stages) or 15 min (at all other times). This discharge was analyzed to elucidate fracture flow and colloid transport in response to rainfall events. Colloid concentrations were observed to increase quickly once rainfall started (~15–90 min) and reached peak values of up to 188 mg/L. Interestingly, maximum colloid concentration occurred prior to the arrival of flow discharge peak (i.e. maximum colloid concentration was observed before saturation of the soil layer). Rainfall intensity (rather than its duration) was noted to be the main factor controlling colloid response and transport. Dissolved organic carbon concentration and $\delta^{18}O$ dynamics in combination with soil water potential were used to apportion water sources of fracture flow at different stages. These approaches suggested the main source of the colloids discharged to be associated with the flushing of colloids from the soil mesopores and macropores. Beyond the scientific interest of colloid mobilization and transport at the field scale, these results have important implications for a region of about 160,000 km^2 in southwest China that featured similar hydrogeologic settings as the experimental site. In this agriculture-dominated area, application of pesticides and fertilizers to farmland is prevalent. These results highlight the need to avoid such applications immediately before rainfall events in order to reduce rapid migration to groundwater via fracture flow in either dissolved form or in association with colloids.

1. Introduction

Colloids are operationally defined as particles with diameters ranging from a few nanometers to several micrometers (Masciopinto et al., 2008; Zhang et al., 2012). They are ubiquitous in subsurface geologic media. Transport of colloids in the subsurface has gained much attention during the last two decades. It has been well recognized that colloids can potentially act as transport vehicles for various contaminants (e.g., radionuclides, heavy metals, pesticides, microbial pathogens) due to their specific surface properties and long-distance transport potentials (e.g., Baek and Pitt, 1996; Gooddy et al., 2007; Kretzschmar et al., 1999; Tang and Weisbrod, 2009, 2010). Colloid transport may not only facilitate the migration of contaminants in colloid-associated forms in porous or fractured media, but also may change soil structure and flow paths (e.g., El-farhan et al., 2000; Majdalani et al., 2008; Weisbrod et al., 2002).

Transport of colloids and colloid-associated contaminants through porous or fractured media at the column or core scale has been widely studied in the laboratory (e.g., Kretzschmar et al., 1999; McCarthy et al., 2002; Mohanty et al., 2013; Mondal and Sleep, 2012; Zhang et al., 2010; Zhuang et al., 2009; Zvikelsky and Weisbrod, 2006). However, much fewer studies report experiments undertaken under field conditions and at large scales (El-farhan et al., 2000; Masciopinto et al., 2008; McKay et al., 1993; Shevenell and McCarthy, 2002; Vilks et al., 1997). High spatial heterogeneities and flow/transport pathway complexities

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could create significant differences between laboratory observations and actual field transport of colloids (Becker et al., 1999; McKay et al., 2000; Weisbrod et al., 2002; Zhang et al., 2012). In the limited number of field-scale colloid transport studies that have been published, focus has, for the most part, been related to the breakthrough of model colloids and conservative tracers injected under transient or steady conditions. For example, faster transport of various colloidal particles (e.g., fluorescent latex microspheres, colloid-sized bacteriophages) as compared with non-reactive solutes (e.g., Br^-, ^{18}O, ^2H) was observed in fractured rocks (Becker et al., 1999; McKay et al., 1993, 2000). Flow rate and ionic strength variation induced by rainfall-infiltration was expected to mobilize the previously retained colloids (McKay et al., 2000). Vilks et al. (1997) demonstrated that silica colloids could be transported over 17 m in fractured granite at the field-scale and that fracture geometry has a great effect on colloid transport. Only a few published studies report on natural colloid transport. The dominance of macropores in soil particle (1–50 μm) transport has been corroborated by Ryan et al. (1998) for macroporous soils in the field. Lægdsmand et al. (1999) also reported that colloids were primarily mobilized through soil macropores in large soil monoliths. Colloid-facilitated transport of various contaminants has been associated with macroporous soils and fractured media. Buddemeier and Hunt (1988) reported colloid-facilitated transport of radionuclides (e.g., Ce, Eu, Mn, Co) through rock fractures at nuclear waste landfill while Gooddy et al. (2007) observed colloid-facilitated transport of pesticide (diuron) through fractured carbonate aquifer. Weisbrod et al. (2002) investigated the temporal variation of particle detachment from shallow fractured chalk in response to tap water percolation at constant head. They reported a large concentration of particles to be detached from the fractures in the first few hours of flooding and decrease in their concentration over time. This observation was repeated in a cyclic pattern in each infiltration event that followed a long dry period. In a deep fractured zone (40–80 m below the land surface) at the Yucca Mountain site, the amount of colloidal particles in natural seepage water was found to be related to solution chemistry, directly influenced by recharge from previous large storms (Cizdziel et al., 2008). Vadose zone flow through a fracture at a karst aquifer in the Swiss Jura Mountains was dynamically analyzed for water percolation and response of particle transport following rainfall events or artificial irrigations (Pronk et al., 2009). However, to the best of our knowledge, no large scale field investigations have been conducted to explore the transport dynamics of naturally released colloids within subsurface fracture flow and their relationships to rainfall intensities.

A vast hilly region (160,000 km^2) of Sichuan in the upper reaches of Yangtze River is characterized by thin purple soil (an entisol according to USDA soil taxonomy) that overlies a fractured calcareous mudrock. The purple soil is known to be very vulnerable to water erosion and poorly aggregated (Li et al., 1991). These circumstances underpin the opportunity for particle detachment and colloid transport. Abundant macropores and fractures develop well in the purple soil and mudrock layer, respectively. Field plot-scale (8 m in length, 4 m in width) experiments were conducted in this region to identify preferential flow and transport paths (Zhao et al., 2013a). Vertical subsurface flow in fractured mudrock was found to be the dominant flow pattern on sloping farmland during rain events (Zhao et al., 2013a). However, there have been no studies regarding the mechanism of subsurface colloid release and transport in this region. Given that the shallow groundwater is the main drinking water source in this region, understanding flow and colloid transport in response to natural rainfall is of the utmost relevance with respect to water safety.

The objectives of the present study were to: (1) capture the dynamic response of subsurface flow and colloid transport to various rainfall events on a representative sloping farmland; (2) relate the governing mechanisms for subsurface flow and colloid transport in this natural setting to intensity and duration of rainfall events.

2. Materials and methods

2.1. Site description

The study site, Yanting, lies in the hilly central Sichuan, southwest China and is featured by the readily erodible purple soil (entisol) overlying fractured mudrock and impermeable sandstone (Fig. 1). The soil and parent rock are similar in mineralogical composition. They contain about 64% of primary minerals such as quartz and feldspar, and the main clay minerals are hydrous mica and montmorillonite (Jiang and Li, 1995). The $CaCO_3$ content is about 10% and 17% in the soil and mudrock, respectively (Jiang and Li, 1995). The annual mean precipitation is 826 mm, with 85% occurring in summer (May–September) (Zhao et al., 2013a). The experimental field is a farmland with an average slope of 6°, located within the Yanting Agro-ecological Experimental Station of Purple Soil (31°16′N, 105°28′E), Chinese Academy of Sciences. On the experimental slope, the thickness of soil layer varied from 20 cm on the upper slope to about 60 cm on the lower slope. The soil showed little evidence of pedogenic horizon development. The shallow soil layer exhibits relatively high saturated hydraulic conductivity in the range of 37–127 mm/h with macroporosity contributing over 87% to flow (Wang, 2013). In this paper, soil macropores and mesopores are defined as water–filled pores at pressure heads between 0 cm and −10 cm, and between −10 cm and −100 cm, respectively. The mudrock (2.1–4.8 m in thickness) beneath the shallow soil layer is heavily weathered with visible discrete fine fractures that have developed within it. Maize and wheat are planted in rotation on the slope without irrigation and the thickness of the plough layer is about 15 cm. The only external water recharge comes from rainfall events.

2.2. Experimental design

2.2.1. Field monitoring and sampling scheme

The experimental plot was hydrologically isolated from the surrounding geologic formations with cement partition walls inserted into the sandstone layer on all four sides of the plot. The underground depths of the partition walls vary from approximately 5.5 m on the upper slope to 3.2 m on the lower slope; these depths correspond to the depths of the of the sandstone layer. A trench (30 m long, 3.5 m deep and 3 m wide) was dug at the lower end of the 50 m long slope. Three conflux grooves were built in the trench with cement and stainless steel pipes for (1) surface flow, (2) interflow discharging at the soil-mudrock interface and (3) fracture flow discharging at the interface of the mudrock and impermeable sandstone underneath, respectively (Fig. 1). The three flow components were monitored for discharge rate and sampled at the outlets of the conflux grooves respectively. However, only the results of the subsurface fracture flow, which was the dominant flow type on the slope and therefore the focus of this study, are presented and analyzed in this article.

Rainfall (amount and duration) was recorded at a 15-min interval by an automatic tipping bucket installed outside and near the mid-slope side wall of the sloping farmland plot. Fracture flow discharge was recorded by a customized tipping bucket gauge installed at the outlet of the conflux groove at a 15-min interval. Two tensiometers (T4e, UMS, München, Germany) were installed at depth of 15 and 45 cm, respectively, at the lower end of the slope to measure soil water potential every 15 min. The tipping