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Transport of *E. coli* D21g with runoff water under different solution chemistry conditions and surface slopes

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SUMMARY

Tracer and indicator microbe runoff experiments were conducted to investigate the influence of solution chemistry on the transport, retention, and release of *Escherichia coli* D21g. Experiments were conducted in a chamber (2.25 m long, 0.15 m wide, and 0.16 m high) packed with ultrapure quartz sand (to a depth of 0.10 m) that was placed on a metal frame at slopes of 5.6%, 8.6%, or 11.8%. Runoff studies were initiated by adding a step pulse of salt tracer or D21g suspension at a steady flow rate to the top side of the chamber and then monitoring the runoff effluent concentrations. The runoff breakthrough curves (BTCs) were asymmetric and exhibited significant amounts of concentration tailing. The peak concentration levels were lower and the concentration tailing was higher with increasing chamber slope because of greater amounts of exchange with the sand and/or extents of physical nonequilibrium (e.g., water flow in rills and incomplete mixing) in the runoff layer. Lower amounts of tailing in the runoff BTC and enhanced D21g retention in the sand occurred when the solution ionic strength (IS) was 100 mM NaCl compared with 1 mM NaCl, due to compression of the double layer thickness which eliminated the energy barrier to attachment. Retained cells were slowly released to the runoff water when the IS of the runoff water was reduced to deionized water. The amount and rate of cell release was greatest at the highest chamber slope, which controlled the amount of exchange with the sand and/or the extent of physical nonequilibrium in the runoff layer, and the amount of retained cells. The observed runoff BTCs were well described using a transient storage model, but fitted parameters were not always physically realistic. A model that accounted for the full coupling between flow and transport in the runoff and sand layers provided useful information on exchange processes at the sand surface, but did not accurately describe the runoff BTCs which were influenced by physical nonequilibrium in the runoff layer.

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1. Introduction

Microorganism contamination of surface water resources is common in the United States (USEPA, 1997; Abbaszadegan et al., 2003; Borchardt et al., 2003), and has frequently been implicated in water- and food-borne disease outbreaks (Centers for Disease Control and Prevention, 1998; USFDA, 1998; Gerba and Smith, 2005; Pachepsky et al., 2011). High concentrations of pathogens, indicator microorganisms, and other colloids can be rapidly transported from agricultural fields, urban environments, and hillslopes to streams or locations of surface water storage by runoff water (Rahe et al., 1978; Heathwaite et al., 2005; Haygarth et al., 2006). Runoff has been reported to be the primary transport route for pathogen dissemination at the hillslope scale (Tyrrel and Quinton, 2003; Jamieson et al., 2004; Dorner et al., 2006). An understanding of processes that influence the transport of pathogens and indicator microorganisms in runoff water is therefore needed to assess and mitigate risks of microbial contamination of surface water supplies to human health.

As pathogens are transported with runoff water they undergo exchange with the soil surface. Exchange of solute and microorganisms to/from runoff water and the soil surface can be very complex, depending on advection, dispersion, and reactions (Bradford et al., 2013). In particular, advective exchange with the soil surface (infiltration and exfiltration) will depend on the surface water boundary conditions, the surface topography, the initial soil water status, hydraulic properties of the soil, soil structure, and subsurface

Abbreviations: DI, deionized; DLVO, Derjaguin–Landau–Verwey–Overbeek; IS, ionic strength.

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heterogeneity. Spatial and temporal variations in these factors can produce conditions of dynamic exchange between runoff water and the soil surface (Wörman et al., 2007). Depression and obstruction storage of surface water will also produce relatively immobile or stagnant zones of water connected to the mean runoff water. Such stagnant zones include pools and eddies along runoff channels, water isolated behind rocks, gravel or vegetation, or relatively inaccessible water due to uneven surface topography (Panday and Huyakorn, 2004). Diffusive exchange will occur between runoff water and such stagnant zones (Wallach and van Genuchten, 1990; Govindaraju, 1996; Wallach et al., 2001). Rain drop impact and erosion of contaminated soils and sediments are other mechanisms of pathogen exchange at the soil surface (Gao et al., 2004, 2005; Pachepsky and Shelton, 2011).

Pathogens at the soil surface may interact with soil particles and/or vegetation and become retained, and thereby diminish their transport with runoff water. A multitude of physical, chemical, and biological processes will influence the retention of microorganisms at the soil surface. For example, microbe retention in soil and sediment is known to depend on soil properties such as surface roughness, grain size and distribution, soil structure, mineralogy, and surface charge; properties of the water such as the water velocity, the water saturation, and the solution pH, ionic strength (IS), and composition; and properties of the microorganisms such as the size and surface charge, surface macromolecules, growth stage, motility, and biofilm (Ginn et al., 2002; Harvey and Harms, 2002; Foppen and Schijven, 2006; Tufenkji et al., 2006; Bradford et al., 2013). Temporal changes in these same factors may produce pathogen release. For example, a decrease in the solution IS will result in an expansion of the double layer thickness and an increase in the magnitude of the surface potential of the soil and microbe (Israelachvili, 1992; Elimelech et al., 1995) that will reduce the depth of the secondary minimum and promote microbe release (Lenhart and Saiers, 2003; Grolimund and Borkovec, 2006; Tosco et al., 2009; Bradford et al., 2012). Runoff water may subsequently become contaminated as a result of diffusion and exfiltration of released pathogens.

Most microbial runoff studies to date have examined the efficacy of vegetated buffer strips to remove pathogens (Atwill et al., 2002; Tate et al., 2004; Trask et al., 2004; Kouznetsov et al., 2007; Guber et al., 2007, 2009; Guzman et al., 2010; Fox et al., 2011; Wu et al., 2011; Yu et al., 2012, 2013). Wide variations in the removal efficiency of vegetative buffer strips have been reported in the literature (Fajardo et al., 2001; Koelsch et al., 2006; Mawdsley et al., 1995; Pachepsky et al., 2006; Tate et al., 2004); e.g., ranging from almost none to complete removal. This variability in performance is likely due to differences in design and operation of vegetative buffer strips, and incomplete characterization of factors influencing exchange with the soil surface. Fox et al. (2011) reported that *Escherichia coli* mass reductions in runoff water were highly correlated with the amount of water infiltration. However, only limited research has addressed the issue of microbial exchange between runoff water and the soil surface. Muirhead et al. (2006) reported that the transport behavior of a conservative tracer (bromide) and *E. coli* in runoff water was almost identical. Yu et al. (2011) reported similar results for kaolinite and bromide in runoff water. However, these studies did not consider the confounding effects of changes in solution chemistry on cell/kaolinite retention and release in the soil. Yu et al. (2013) reported greater retention of latex microspheres to surface vegetation occurred at higher IS and for larger particles. Ferguson et al. (2007) found that larger microorganisms (*Cryptosporidium* and *E. coli*) were more efficiently removed from runoff water than smaller microbes (coliphage PRD1) in field studies. Furthermore, previous studies have not yet modeled the full dynamics of water flow and microbial transport in runoff water and soil. Most modeling studies only consider runoff and transport at the soil surface, and approximate exchange between runoff water and the soil using simplified terms for infiltration and/or diffusive exchange (Guber et al., 2009; Yu et al., 2011, 2012; Wu et al., 2011; van Genuchten et al., 2013).

The objective of this research was to study the effects of slope and solution IS on the transport, exchange, retention, and release of an indicator microorganism (*E. coli* D21g) in runoff water. In particular, different amounts of advective exchange between the runoff water and the soil surface were achieved by varying the slope (5.6%, 8.6%, and 11.8%) of a chamber packed with fine sand. The IS of the runoff water was varied (1 and 100 mM NaCl) to obtain a wide range of cell retention and release (deionized water) conditions. Experiments were simulated using the transient storage model and a model that describes the full coupling of flow and transport (tracer and *E. coli* D21g) between runoff water and the sand in the chamber.

2. Materials and methods

2.1. Electrolyte solutions and porous media

Electrolyte solutions were prepared using deionized (DI) water (pH = 5.8) to achieve 0, 1, or 100 mM NaCl solutions. Ultrapure quartz sand (Iota Quartz, Unimin Corp. NC) was employed as a porous medium in the runoff experiments discussed below. The sand was thoroughly rinsed with DI water to eliminate background fines from the sand before use. The median grain size of this sand was measured to be 238 μm (standard deviation of 124 μm) with a laser scattering particle size and distribution analyzer (Horiba LA 930). The saturated conductivity (K_s) of the sand in a packed column was determined to be 0.57 cm min^{-1} using Darcy's law and measurements of the hydraulic gradient during steady-state water flow. The porosity of the packed column was calculated to be 0.44 from the measured bulk density (1.49 g cm^{-3}) and an assumed specific density of the sand (2.65 g cm^{-3}).

2.2. E. coli D21g

Experiments discussed below employed pure cultures of *E. coli* D21g, which is a gram-negative, nonmotile bacterial strain that produces minimal amounts of lipopolysaccharides and extra-cellular polymeric substances. *E. coli* D21g was grown 24 h before initiating experiments. A single colony of *E. coli* D21g was inoculated into 1000 mL of Luria–Bertani media containing 30 μg mL^{-1} gentamycin and incubated with shaking at 37 °C overnight. The bacterial suspension was centrifuged and rinsed three times before diluting the concentrated suspension into the desired electrolyte solution. Influent and effluent concentrations of *E. coli* D21g were determined using a spectrophotometer (Unico UV-2000, United Products & Instruments, Dayton, NJ) at a wavelength of 600 nm and a calibration curve. The average optical density at 600 nm for the influent cell suspension was 0.077, which corresponds to an input concentration (C_o) of approximately $1.1 * 10^6$ cells mL^{-1}.

2.3. Interaction energy calculations

The zeta potential of *E. coli* D21g and crushed ultrapure quartz sand in the various solution chemistries was determined from measured electrophoretic mobilities using a ZetaPALs instrument (Brookhaven Instruments Corporation, Holtsville, NY) and the Smoluchowski equation. The size of *E. coli* D21g in the various solution chemistries was measured using the laser scattering particle size and distribution analyzer. The total interaction energy of *E. coli* D21g upon approach to the quartz sand under the various