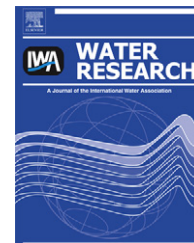


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Impact of dissolved organic matter on colloid transport in the vadose zone: Deterministic approximation of transport deposition coefficients from polymeric coating characteristics

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ABSTRACT

Although numerous studies have been conducted to discern colloid transport and stability processes, the mechanistic understanding of how dissolved organic matter (DOM) affects colloid fate in unsaturated soils (i.e., the vadose zone) remains unclear. This study aims to bridge the gap between the physicochemical responses of colloid complexes and porous media interfaces to solution chemistry, and the effect these changes have on colloid transport and fate. Measurements of adsorbed layer thickness, density, and charge of DOM-colloid complexes and transport experiments with tandem internal process visualization were conducted for key constituents of DOM, humic (HA) and fulvic acids (FA), at acidic, neutral and basic pH and two CaCl₂ concentrations. Polymeric characteristics reveal that, of the two tested DOM constituents, only HA electrosterically stabilizes colloids. This stabilization is highly dependent on solution pH which controls DOM polymer adsorption affinity, and on the presence of Ca⁺² which promotes charge neutralization and inter-particle bridging. Transport experiments indicate that HA improved colloid transport significantly, while FA only marginally affected transport despite having a large effect on particle charge. A transport model with deposition and pore-exclusion parameters fit experimental breakthrough curves well. Trends in deposition coefficients are correlated to the changes in colloid surface potential for bare colloids, but must include adsorbed layer thickness and density for sterically stabilized colloids. Additionally, internal process observations with bright field microscopy reveal that, under optimal conditions for retention, experiments with FA or no DOM promoted colloid retention at solid-water interfaces, while experiments with HA enhanced colloid retention at air-water interfaces, presumably due to partitioning of HA at the air-water interface and/or increased hydrophobic characteristics of HA-colloid complexes.

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1. Introduction

Dissolved organic matter (DOM) plays a prominent role in many soil processes and is ubiquitous in soils; high concentrations are found in manure or wastewater sludge amended lands. Humic acid (HA) and fulvic acid (FA) are principal constituents of soil, aquatic, sewage sludge, and manure DOM (Schnitzer, 1972; Thurman, 1985). The structural properties of the moieties that make up DOM in soil environments have been explored by several investigators; reporting (as general consensus) that DOM molecules behave as flexible entities that can swell and shrink in response to changes in pH and ionic strength (Avena et al., 1999; Benedetti et al., 1996; Duval et al., 2005; Hosse and Wilkinson, 2001). The amphiphilic character (i.e., presence of hydrophobic and hydrophilic moieties) of DOM has been reported by a number of studies (Guetzloff, 1994; Lenhart and Saiers, 2004; Ma et al., 2007; von Wandruszka, 2000) and used to explain its high surface reactivity and adsorptive fractionation to solid-water and air-water interfaces (Chi and Amy, 2004; Lenhart and Saiers, 2004; Ma et al., 2007). Greater affinity of larger, more hydrophobic DOM components for mineral surfaces and air-water interfaces is of primary importance for contaminant flux through the vadose zone (Lenhart and Saiers, 2004; Meier et al., 1999), as this unsaturated soil region is the critical connection that buffers deep groundwater from surface and shallow contaminants.

Physicochemical interactions between DOM and contaminants have received considerable attention in recent years. Numerous investigations have demonstrated that even small amounts of DOM greatly increase the mobility of colloid-associated contaminants (e.g., radionuclide plutonium, americium, thorium, and radium; phosphorus; hydrophobic organic compounds; uranium(IV)/(VI); carbon nanotubes; and lead) (Flury and Qiu, 2008; Granger et al., 2007; Jaisi et al., 2008; Marley et al., 1993; Mibus et al., 2007; Sen and Khilar, 2006; Tang and Weisbrod, 2009) and colloid-sized pathogens (e.g., *Escherichia coli*, *Cryptosporidium parvum* oocysts, *Giardia*, and bacteriophage PRD1) (Abudalo et al., 2005, 2010; Bradford et al., 2006; Foppen et al., 2008) through hydrologic pathways. Laboratory batch kinetic and isotherm experiments have also been conducted to explore the interactions between DOM and colloidal particles. Results from these experiments indicate that DOM increases the stability of colloid and nanoparticle suspensions in the presence of electrolytes through electrostatic and/or steric stabilization by way of adsorption onto colloid surfaces (Akbour et al., 2002; Chen and Elimelech, 2007; Heidmann et al., 2005; Kretzschmar et al., 1998; Pefferkorn, 2006). Moreover, complexation of surface functional groups with non-indifferent ions in solution is a widely recognized process (Amirbahman and Olson, 1995; Chen and Elimelech, 2007; Chen et al., 2006) that could significantly affect the stability and therefore the transport of colloids suspended in DOM rich solutions.

A number of investigations systematically examined the impact of DOM on colloid mobility in saturated porous media in terms of pore water velocity and deposition kinetics (Akbour et al., 2002; Jaisi et al., 2008; Kretzschmar et al., 1997). The effects of mono- vs. divalent cation concentrations (Jaisi

et al., 2008), and ionic strength on attachment efficiency have been evaluated (Franchi and O'Melia, 2003; Kretzschmar and Sticher, 1997), as well as charge reversal by organic matter adsorption (Kretzschmar and Sticher, 1997). Both natural and well characterized porous media have been used in research spanning acidic and neutral pore water pH ranges (Akbour et al., 2002; Franchi and O'Melia, 2003; Jaisi et al., 2008; Kretzschmar et al., 1997; Kretzschmar and Sticher, 1997). However, only limited studies have explored the effect of DOM and pH on colloid transport in unsaturated porous media (Tang and Weisbrod, 2009) and the effect of colloid transport in alkaline DOM rich conditions (Harvey et al., 2010). This range of solution chemistry is of high relevance for soils amended with lime-treated manure as a commonly used pathogen inactivation treatment. Two general consensus about the presence of air phases are that interfaces with air in unsaturated porous media promote colloid retention, and that organic matter of hydrophobic character preferentially fractionates to the air-water interface. Thus, it is of critical importance to understand the effect that DOM has on colloid transport in unsaturated soils; particularly if adsorbed amphiphilic DOM may provide hydrophobic characteristics to the interfaces and colloid surfaces it adsorbs onto.

This study aims to experimentally bridge the gap between the physicochemical changes of DOM-colloid complexes and porous media interfaces and the effect that these systematic changes have on the transport of colloids in unsaturated soils. These objectives will be achieved by: (i) directly measuring the changes in surface charge, adsorbed layer thickness and density of organic matter-colloid complexes under acidic, neutral, and basic solution pH in the presence and absence of CaCl₂, (ii) assessing with column experiments the effects that HA and FA have on colloid transport at the solution chemistries listed above, (iii) simultaneous internal observation of the dominant pore scale retention sites for each set of conditions, and (iv) mathematical modeling of the transport behavior of organic matter-colloid complexes to relate deposition coefficients with changes in solution composition.

2. Materials and methods

2.1. Preparation of materials

In order to meet the objective of discerning the specific steric characteristics that increase the stability of organic matter-colloid complexes (e.g., thickness of adsorbed organic matter layer and uniform adsorption of DOM onto colloid surfaces), the use of uniform and spherical colloids was essential. As such, calibration grade polystyrene and carboxylated spheres of 24 nm diameter (Bangs Laboratories Inc.; Fishers, IN) were used to measure the adsorbed layer thickness. These spheres were selected because of their exceptional size uniformity that allowed the measurement of changes in size at the nanometer scale. Similar surfactant-free, red-dyed, polystyrene and carboxylated spheres of 2.6 μm diameter (Magsphere, Pasadena, CA) were used for all other measurements. The larger red colloids were chosen because they permitted excellent visualization of individual colloids against the porous medium with Bright Field Microscopy (BFM).

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