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Cotransport of clay colloids and viruses through water-saturated vertically oriented columns packed with glass beads: Gravity effects

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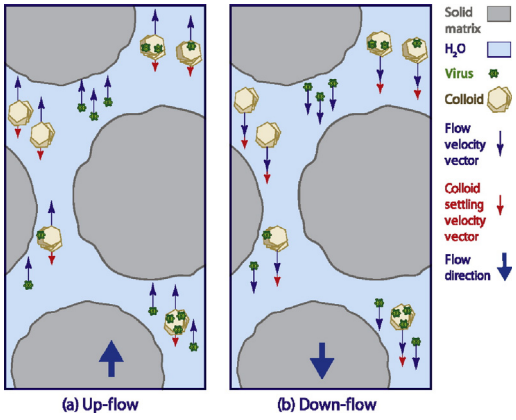
HIGHLIGHTS

- Gravity affects viruses and clay colloids cotransport in porous media.
- Colloids can facilitate or hinder the transport of viruses in porous media.
- Virus attachment in the presence of colloids is greater for up-flow than down-flow.

GRAPHICAL ABSTRACT



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ABSTRACT

The cotransport of clay colloids and viruses in vertically oriented laboratory columns packed with glass beads was investigated. Bacteriophages MS2 and ΦX174 were used as model viruses, and kaolinite (KGa-1b) and montmorillonite (STx-1b) as model clay colloids. A steady flow rate of Q = 1.5 mL/min was applied in both vertical up (VU) and vertical down (VD) flow directions. In the presence of KGa-1b, estimated mass recovery values for both viruses were higher for VD than VU flow direction, while in the presence of STx-1b the opposite was observed. However, for all cases examined, the produced mass of viruses attached onto suspended clay particles were higher for VD than VU flow direction, suggesting that the flow direction significantly influences virus attachment onto clays, as well as packed column retention of viruses attached onto suspended clays. KGa-1b hindered the transport of ΦX174 under VD flow, while STx-1b facilitated the transport of ΦX174 under both VU and VD flow directions. Moreover, KGa-1b and STx-1b facilitated the transport of MS2 in most of the cases examined except of the case where KGa-1b was present under VD flow. Also, the experimental data were used for the estimation of virus surface-coverages and virus surface concentrations generated by virus diffusion-limited attachment, as well as virus attachment due to sedimentation. Both sedimentation and diffusion limited virus attachment were higher for VD than VU flow, except the case of MS2 and STx-1b cotransport. The diffusion-limited attachment was higher for MS2 than ΦX174 for all cases examined.

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1. Introduction

Numerous experimental and theoretical studies published in the literature have focused on factors that govern colloid and biocolloid (e.g. viruses, bacteria) transport in fractured and porous media (Zhuang and Jin, 2003; Bradford et al., 2006; Torkzaban et al., 2007; Kim et al., 2009; Chrysikopoulos et al., 2010, 2012; Syngouna and Chrysikopoulos, 2015; Sen, 2011; Shen et al., 2012; Katzourakis and Chrysikopoulos, 2014, 2015; Seetha et al., 2015; Kokkinos et al., 2015), have examined the role of velocity on colloid transport and deposition, in conjunction with the influence of either the particle size (e.g. Bradford et al., 2006; Chrysikopoulos and Katzourakis, 2015), or the ionic strength (e.g., Tong et al., 2008; Tosco et al., 2009; Mitropoulou et al., 2013; Torkzaban et al., 2015). Other factors that have been evaluated include the role of roughness and flow direction (Yoon et al., 2006; Bradford and Torkzaban, 2013), collector size (Xu et al., 2006; Syngouna and Chrysikopoulos, 2011), pore geometry or grain angularity (Tong and Johnson, 2006; Ma and Johnson, 2010), and chemical heterogeneity of the porous media (Bradford and Torkzaban, 2012). Moreover, the presence of colloids suspended in the aqueous phase has been shown to either enhance or hinder the transport of organic and inorganic pollutants (Kretzschmar et al., 1999; Walshe et al., 2010; Syngouna and Chrysikopoulos, 2013).

The effect of flow direction on colloid and biocolloid transport in porous media has received relatively minor attention (Ma et al., 2009; Chrysikopoulos and Syngouna, 2014). The flow direction employed in typical contaminant transport experimental investigations is horizontal (Silliman et al., 2001; Syngouna and Chrysikopoulos, 2011, 2013; Vasiliadou and Chrysikopoulos, 2011), downward (Anders and Chrysikopoulos, 2009; Chrysikopoulos et al., 2010; Xu et al., 2006), or upward (Bradford et al., 2006; Tong and Johnson, 2007). The upward flow direction is often selected in order to minimize air entrapment. Previous experimental observations have revealed that flow direction influences colloid transport in porous media showing greater rate of particle deposition for up-flow than for down-flow direction and suggesting that gravity was a significant driving force for colloid deposition (Chrysikopoulos and Syngouna, 2014). However, the effect of flow direction on the cotransport of clay colloids and viruses has not been previously explored.

The present study examined the effect of flow direction on the cotransport of clay colloids and viruses in vertical water-saturated columns packed with glass beads. A steady flow rate was applied in both vertical up (VU) and vertical down (VD) directions. Bench scale experiments were performed to investigate the interactions between viruses and clays during their simultaneous transport (cotransport) in porous media. Also the synergistic effects of suspended clay colloids and flow direction on the attenuation and transport of viruses in porous media was examined. Furthermore, virus diffusion-limited attachment and virus attachment by sedimentation were evaluated for all the cotransport experiments conducted in this study.

2. Materials and methods

2.1. Bacteriophages and assay

The bacteriophage MS2 (F-specific single-stranded RNA phage with effective particle diameter ranging from 24 to 26 nm) has been recommended as a surrogate for poliovirus due to similarities in size, and has been employed as a conservative tracer for enteric virus transport, because MS2 attachment onto the majority of soil types is low compared to many other viruses (Jin and Flury, 2002; Schjiven et al., 1999). Moreover, the bacteriophage ΦX174 (somatic single-stranded DNA phage with effective particle diameter ranging from 25 to 27 nm) has been recommended as an indicator for enteric viral pathogens (Gantzer et al., 1998). Both bacteriophages are infecting *Escherichia coli*, and were assayed by the double-layer overlay method (Adams, 1959), as outlined by Syngouna and Chrysikopoulos (2011).

Each initial virus concentration used in this study, collected from the same virus stock solution was diluted with sterile distilled deionized water (ddH_2O), which was purified with a Milli-Q UV plus water purification system (Millipore Corp., Massachusetts). The resulting viral suspension was close to neutral. For the separation of viruses attached onto clay colloids from suspended viruses in the liquid phase, 0.3 mL of the density gradient separation reagent Histodenz (60% by weight, Axis-Shield PoC AS Company, Norway) was added to 2 mL of the liquid sample (Vasiliadou and Chrysikopoulos, 2011; Jiang et al., 2007; Rong et al., 2008), the mixture was centrifuged at 2000×g for 30 min so that the supernatant was free of clay colloids.

The suspension of unattached viruses in the supernatant was pipetted out and the suspended viruses were determined. The absence of clay colloids in the supernatant was verified by a UV–vis spectrophotometer (UV-1100, Hitachi) at a wavelength of 280 nm. The concentration of attached viruses was determined by subtracting the mass of viruses that remained in suspension from the initial virus concentration in each sample. Because only viable viruses were measured in the water samples, it was important to exclude the effect of virus inactivation when evaluating interactions of viruses with clay colloids under the present experimental conditions. Previous batch inactivation experiments under identical experimental conditions, in the presence and absence of clays, suggested that no significant virus inactivation is expected during the experimental time period (Chrysikopoulos and Syngouna, 2012; Bellou et al., 2015). Although the inactivation rates of the viruses used in this study are relatively small, MS2 inactivation rate of 4.2×10^{-5} min^{-1} was reported to be more than two times larger than 2.0×10^{-5} min^{-1} that of ΦX174 (Syngouna and Chrysikopoulos, 2011). Furthermore, the inactivation rate of MS2 was reported to be greater than that of ΦX174 in the presence of quartz sand under static and dynamic batch conditions at different temperatures (Chrysikopoulos and Aravantinou, 2012). Therefore, the difference in the inactivation rate coefficients between MS2 and ΦX174is expected to yield smaller MS2 breakthrough concentrations than ΦX174. Note that inactivation of suspended viruses in water saturated porous media is controlled by the physicochemical characteristics of viruses (Yamagishi and Ozeki, 1972), formation temperature (Yates and Yates, 1988), and time (Sim and Chrysikopoulos, 1996; Anders and Chrysikopoulos, 2006). Certainly, the inactivation rates of liquid-phase and attached viruses should not be assumed equal (Chrysikopoulos and Sim, 1996; Sim and Chrysikopoulos, 1999). Moreover, inactivation of clay-attached viruses can either be enhanced due to distortion and unfolding of protein structure caused by strong electrostatic attraction, or reduced due to virus protection provided by the clay particles (Schijven and Hassanizadeh, 2000; Ryan et al., 2002).

2.2. Clays

The clays used in this study were kaolinite (KGa-1b, well-crystallized kaolin, from Washington County, Georgia) (Pruett and Webb, 1993), and montmorillonite (STx-1b, Ca-rich montmorillonite, Gonzales County, Texas), purchased from the Clay Minerals Society (Columbia, USA). KGa-1b has specific surface area (SSA) of 10.1 m^2/g, as evaluated by the Brunauer–Emmet–Teller (BET) method, and cation exchange capacity (CEC) of 2.0 meq/100 g (van Olphen and Fripiat, 1979). STx-1b has a SSA of 82.9 m^2/g (Sanders et al., 2010), and assuming that the characteristics of STx-1b are comparable to those of STx-1, which is the previous batch of montmorillonite from the same area, its CEC is 84.4 meq/100 g (van Olphen and Fripiat, 1979). The <2 μm colloidal fraction, used in the transport experiments, was separated by sedimentation and was purified following the procedure described by Rong et al. (2008). It should be noted that the treated clays (purified colloidal fraction) are smaller than the untreated particles, and thus is reasonable to assume that the treated clays are expected to have higher SSA values