Removal of Pathogenic Microorganisms during Riverbank Filtration

Project Scope

Riverbank filtration holds promise as a relatively simple and low-cost way to remove particulates and microorganisms from surface water and make subsequent disinfection treatment easier. In this method, extraction wells are placed deep within riverbank (alluvial) sediments, where they collect river water that filters through the sediments. The effectiveness of this method—i.e., the degree to which microbial pathogens are removed from the collected water—depends greatly on physical processes and aquifer sediment properties (e.g., grain size and distribution) which varies greatly among sites.

This grant characterized the physical processes and sediment properties that affect pathogen transport through riverbank aquifer sediments. The research will improve predictions of the effectiveness of riverbank filtration, which will help minimize expensive site-specific testing to characterize microbial transport potential.

The major objective of this grant was to develop a model of *Cryptosporidium parvum* oocyst transport in porous media that can accommodate the physical and geochemical variety present in riverbank aquifers. The researchers tested the following:

1. effects of physical and geochemical heterogeneity of porous media on oocyst transport in flow-through columns and an intermediate-scale two-dimensional aquifer tank,
2. effect of organic matter on oocyst transport in a geochemically heterogeneous porous medium,
3. roles of straining and secondary minimum deposition in oocyst removal, and
4. effects of removal mechanisms on the deposition kinetics of microbes.

Grant Title and Principal Investigators

Microbial Pathogen Removal during Riverbank Filtration (EPA Grant #R829010)

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Key Findings and Implications

Analytical Accomplishments:

- Evaluated oocyst transport in a physically and geochemically heterogenous porous medium in packed flow-through columns and an intermediate-scale two-dimensional aquifer tank that simulates riverbank filtration conditions.
- Demonstrated that geochemical or surface charge heterogeneity play an important role in *Cryptosporidium parvum* oocyst transport in porous media.
- Demonstrated oocyst removal by straining (i.e., capture of particles in sediment pores smaller than the particles) is an important result of physical heterogeneity (grain size differences) in porous media.
- Provides data for a model that will address key microbial transport processes in riverbank sediments.

Implications of Research:

- Physical and geochemical heterogeneities of porous media affect microorganism transport and must be accounted for in predicting pathogen removal during riverbank filtration.
- Will help lead to improved understanding of the effectiveness of riverbank filtration in pathogen removal and inform research and future risk management decision-making on the use of riverbank filtration as a water treatment method.

Publications include 5 peer reviewed journal articles.

Project Period: September 2001 to August 2004 (extended through August 2005)

Relevance to ORD's *Drinking Water Research Multi-Year Plan* (2003 Edition)

This project contributes directly to the third of three Long-term Goals for drinking water research: (3) By 2009, provide data, tools and technologies to support management decisions by the Office of Water, state, local authorities, and utilities to protect source water and the quality of water in the distribution system.

The preliminary and future results from this research grant will improve our understanding of the effectiveness of riverbank filtration in pathogen removal, a pre-treatment option that may help prevent outbreaks of waterborne disease and reduce compliance costs. The research provides data about fundamental processes governing pathogen transport in groundwater systems. These data are necessary for the development of predictive models to screen alluvial valley aquifers as candidates for riverbank filtration. Accurate data and predictive tools will support future investment and management decisions regarding the use of riverbank filtration as a water treatment method. For example, this research provides data for EPA's proposed Long-Term 2 Enhanced Surface Water Treatment Rule that allows riverbank filtration as a "prefiltration toolbox" option that water utilities can use to help meet log-removal requirements for *Cryptosporidium parvum*.

The researchers collected data and tested models using an intermediate-scale two-dimensional aquifer tank (4.6 m length × 1.0 m height) and packed flow-through columns.

Project Results and Implications

Grain Size and Surface Charge Heterogeneity Effects on Oocyst and Microsphere Transport: The investigators measured transport of *Cryptosporidium parvum* oocysts and polystyrene latex microspheres (as surrogates for oocysts) in physically and geochemically heterogenous porous media packed into flow-through columns. They examined the effects of grain size and surface charge heterogeneity on microbe and colloid deposition processes and applied these concepts to a larger scale model. Comparisons of model predictions to experimental data indicated that the observed non-exponential deposition behavior of bacteria and virus particles may be attributed to a broad range (i.e., a power-law distribution) of microbial deposition rates. Other mechanisms, such as particle release and blocking by previously deposited microbial particles, were also shown to be potential sources of deviation from the classical filtration theory. These results suggest that monitoring fluid-phase particle concentration is insufficient for accurate characterization of the deposition and transport behavior of microbial particles in saturated porous media. Rather, the shape of the microbial particle retention profile was shown to be a key indicator of the mechanisms controlling microbial deposition and transport.

Geochemical Heterogeneity Effects on Oocyst and Microsphere Transport: The researchers examined next the effect of geochemical heterogeneity of the porous medium on the transport of *C. parvum* oocysts and a virus, the bacteriophage PRD1, in flow-through column experiments. Geochemical heterogeneity was in the form of coatings of ferric oxyhydroxide on quartz grains, which results in positively charged patches within a negatively charged background at solution pH values between the points of zero charge of the ferric oxyhydroxide (about 8) and quartz (about 2.5). For phage PRD1, the collision efficiency (α, the probability of attachment following a microbe-grain collision) was found to increase linearly with the fraction of surface coating (λ), corresponding closely to $\alpha \approx \lambda$. For the oocysts, the collision efficiency also increased with increasing fraction of surface coating, but the slope of the relationship was steeper (i.e., α was greater than λ). Using sand with a four percent surface coating and increasing the pH of the solution to as high as 10 resulted in a decrease in the oocyst collision efficiency. The pH increase presumably reversed the charge of the ferric oxyhydroxide patches from positive to negative. Adding dissolved organic matter at concentrations up to 20 mg L^{-1} reduced the removal of oocysts.

The effects of geochemical heterogeneity on the process of colloid deposition was further examined by conducting experiments with polystyrene latex microspheres of three different sizes in the presence of a simple salt, an anionic surfactant, and at high pH. These experiments revealed the controlling influence of deposition in the "secondary minimum" (i.e., attractive interactions between a colloid and grain that exist at fairly large distances of separation between the colloid and grain under some solution chemistry conditions), and this secondary minimum deposition is the cause of some deviation from the deposition patterns expected under the colloid filtration theory. The experiments in the presence of the anionic surfactant—sodium dodecyl sulfate, which was added to impart a negative change to near-neutrally charged regions of particle and collector surfaces—reduced the deviation from colloid filtration theory by reducing the importance of secondary minimum deposition. High solution pH was found to mask the influence of positively-charged metal oxide impurities on glass bead surfaces and also reduced the deviation from colloid filtration theory. The results demonstrate that both secondary minimum deposition and surface charge heterogeneities contribute significantly to the deviation of colloid deposition processes from colloid filtration theory.

<u>Straining Effects on Oocyst and Microsphere Transport</u>: Recent work has shown that straining, the trapping of particles in pores smaller than the particles, may be an additional important removal mechanism for microorganisms. The investigators conducted oocyst and polystyrene latex microsphere transport experiments using quartz sand and glass bead columns and found that oocyst deposition rates that were higher that expected for the solution chemistry conditions, which suggested that a physical mechanism—straining—must have played a key role in oocyst removal. Supporting experiments conducted using microspheres of varying sizes under very low ionic strength conditions where physicochemical filtration is negligible, clearly indicated that physical straining is an important capture mechanism of microorganisms in subsurface environments. Straining became important for microspheres of 4.1μm diameter (similar to oocysts) in 0.21 mm mean diameter angular quartz sand, a microbe to grain size ratio of 0.002. Using glass beads of 0.23 mm mean diameter as an alternative porous medium, the study also showed that irregularity of sand grain shape contributes considerably to the straining potential of the porous medium. To further examine the effect of physical heterogeneity on microorganism transport, the investigators evaluated the breakthrough of oocysts in well-rounded quartz sands of different grain size, ranging from 0.099 to 2.18 mm. After exploring a variety of experimental techniques intended to detect the grain size at which straining becomes important, it was determined that an energy input (in the form of column vibration) was necessary to definitively identify oocyst removal by straining. Using this technique, the investigators determined that straining of oocysts was significant in quartz grains of less than 0.46 mm, or at a ratio of oocyst diameter to grain diameter of about 0.0001.

<u>Physical and Geochemical Heterogeneity Effects on Oocyst Transport in an Intermediate-Scale Two-Dimensional Aquifer Tank</u>: The investigators simultaneously evaluated the effects of physical and geochemical heterogeneity on oocyst transport in the intermediate-scale two-dimensional aquifer tank. The tank was randomly packed with sands of different grain size (0.12 to 0.86 mm) and different fractions of ferric oxyhydroxide surface coating (0 to 16 percent) in cells of 20 cm length and 5 cm height. The transport of a conservative tracer (nitrate), oocysts, and carboxylated polystyrene latex microspheres of 0.11, 1.0, and 4.0 μm size was measured in vertical arrays of sampling ports at six different transport distances. Oocyst breakthrough was found to be controlled primarily by the physical heterogeneity as most of the oocyst transport occurred along paths dominated by cells of larger grain size. The 4.6 μm microspheres generally followed the same paths at essentially the same rate as the oocysts, but their breakthrough was attenuated to a greater extent. The transport of the smaller microspheres was still being evaluated to examine the effect of size on colloid transport in a heterogeneous medium. Modeling of oocyst and microsphere breakthrough is still ongoing using a model developed by Elimelech and colleagues during previous EPA-funded research. The model has been adapted to include straining and reversible secondary minimum deposition in addition to physicochemical filtration as oocyst and microsphere removal mechanisms.

Investigators

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For More Information

NCER Project Abstract and Reports:

http://cfpub2.epa.gov/ncer_abstracts/index.cfm/fuseaction/display.abstractDetail/abstract/1067/report/0

Peer Reviewed Publications

Tufenkji, N., Ryan, J.N., and Elimelech, M. 2002. The promise of bank filtration. Environmental Science and Technology A Pages 36(21):422A-428A.

Tufenkji, N., Elimelech, M., and Redman, J.A. 2003. Interpreting deposition patterns of microbial particles in laboratory-scale column experiments. Environmental Science and Technology 37(3):616-623.

Tufenkji, N., and Elimelech, M. 2004. Correlation equation for predicting single collector efficiency in physicochemical filtration in saturated porous media. Environmental Science and Technology 38(2):529-536.

Tufenkji, N., Miller, G.F., Ryan, J.N., Harvey, R.W., and Elimelech, M. 2004. Transport of *Cryptosporidium* oocysts in porous media: Role of straining and physicochemical filtration. Environmental Science and Technology 38(22):5932-5938.

Tufenkji, N., and Elimelech, M. 2005. Breakdown of colloid filtration theory: Role of the secondary minimum and surface charge heterogeneities. Langmuir 21(3):841-852.

Abudalo, R.A., Bogatsu, Y.G., Ryan, J.N., Harvey, R.W., Metge, D.W., and Elimelech, M. 2005. The effect of ferric oxyhydroxide grain coatings on the transport of bacteriophage PRD1 and *Cryptosporidium parvum* oocysts in saturated porous media. Environmental Science and Technology (accepted).