Report as of FY2007 for 2006WA147B: "Transport of Colloids in Soils Studied by Geocentrifuge Techniques"

Publications

- Water Resources Research Institute Reports:
  - Flury, Markus and Prabhakar Sharma, 2007, Transport of Colloids in Soils Studied by Geocentrifuge Techniques, State of Washington Water Research Center, Washington State University, Pullman, Washington. State of Washington Water Research Center Report WRR-28, 26 pages.
- Other Publications:
  - Sharma, Prabhakar. 2007. Studying Colloid Transport in Porous Media at Different Acceleration. Oral presentation to the Department of Crop and Soil Science, Washington State University, Pullman, Washington.
  - Sharma, Prabhakar, Markus Flury, and E. Mattson. 2006. Fate and Transport of Colloid in the Subsurface. Poster Presentation at the Annual meeting of Agricultural and Biological Engineers, Portland, Oregon.
  - Sharma, Prabhakar, Markus Flury, and E. D. Mattson, 2006, On the use of Geocentrifuges to Study Colloid Transport in Porous Media, Oral Presentation at the Inland Northwest Research Alliance 2006 Environmental and Subsurface Science Symposium, Moscow, Idaho, September 25-27, 2006.
- Articles in Refereed Scientific Journals:
  - Sharma, Prabhakar; Markus Flury; and E.D. Mattson, Studying Colloid Transport in Porous Media using Geocentrifuge. Submitted to Environment Science Technology (in review).

Report Follows

PROBLEM AND RESEARCH OBJECTIVES

Pharmaceuticals, pathogens, pesticides, and heavy metals often move through soils by the process of colloid or colloid-facilitated transport. Colloids are commonly defined as particles less than 10 μm in diameter that can remain suspended in aqueous solution for considerable amounts of time. Pharmaceuticals, pesticides, and heavy metals are prone to attach to colloids in soils, and pathogenic microorganisms are considered colloids themselves. It is therefore important to understand the mechanisms of colloid transport in soils. The length of time to conduct flow and transport experiments in porous media had led researchers to use centrifuges as tools to evaluate subsurface transport processes. Geocentrifuges are particularly useful to study transport processes in soils, because the flow rates, which are inherently slow in soils, can be sped up considerably. However, there is a need to assure the applicability of geocentrifuges for studying colloid transport. The overall goal of this study was to test the suitability of geocentrifuges to study colloid transport in soils. Specifically, we addressed the following objectives:

- Determine critical centrifugal accelerations as a function of colloid specific density and diameter.
- Evaluate the correction factor in the theoretical relationship for critical acceleration by using geocentrifuges.

This study combines colloid transport theory with laboratory experimentation. First, we developed a theoretical framework to describe colloid transport in geocentrifuges. We rigorously tested the theory by a series of geocentrifuge experiments. Colloid filtration experiments were carried out with representative soil colloids under different centrifugal accelerations.

METHODOLOGY

Objective 1: *Determine critical centrifugal accelerations as a function of colloid specific density and diameter*

Colloid removal from the fluid phase during transport in porous media under favorable attachment conditions can be described by filtration theory (Rajagopalan and Tien, 1976). However, under conditions unfavorable for colloid attachment, filtration theory does not apply. It was observed that colloid concentration profiles in porous media did not follow the exponential decrease expected from filtration theory (Tufenkji and Elimelech, 2004). Another deviation from filtration theory is that under unfavorable conditions, deposition rates of colloids decrease with increasing flow rate (Tong and Johnson, 2006).

In many environmental situations, colloid attachment occurs under unfavorable conditions (Johnson et al., 2005). No theory is currently available to predict colloid deposition under unfavorable conditions. However, it is expected that sedimentation and diffusion play an important role in the deposition process. To assess the effect of acceleration on colloid transport in porous media, we therefore use a theory based on sedimentation and diffusion. We consider the relative importance of sedimentation and diffusion as affected by acceleration.

The root mean square displacement of a colloid by diffusion is (Hiemenz and Ragagopalan, 1997):

$$\bar{x} = \sqrt{2Dt} \qquad (1)$$

where D is the diffusion coefficient, k is the Boltzmann constant, T is absolute temperature, η is the dynamic viscosity, d_c is the colloid diameter, and t is time.

The apparent velocity of diffusion is:

$$v_{diff} = \frac{x}{t} = \sqrt{\frac{2D}{t}} = \frac{2D}{x} \qquad (2)$$

where x is distance. The apparent velocity of diffusion is time- or scale-dependent. With increasing time or distance, the diffusive velocity decreases. The sedimentation velocity of a colloid is given by Stokes law as:

$$v_{sed} = \frac{d_c^2 a(\Delta\rho)}{18\eta} \qquad (3)$$

where a is the acceleration, and $\Delta\rho$ is the density difference between the colloid and the fluid. The random motion of colloids due to diffusion counteracts the linear motion due to sedimentation. In a centrifugation experiment, diffusion is not affected, but sedimentation increases with acceleration, thereby the balance between the two processes changes. We assume that applying a centrifugal acceleration to a colloid transport experiment will not affect the transport results (i.e., colloid deposition) as long as the sedimentation velocity is less or equal to the diffusion velocity multiplied by an empirical factor:

$$v_{sed} \le \xi v_{diff} \qquad (4)$$

The parameter   includes processes other than sedimentation and diffusion that lead to colloid deposition in a porous medium. When the centrifugal acceleration exceeds a certain threshold, sedimentation dominates diffusion. We can derive the relationship between this threshold acceleration a_T as a function of colloid density and colloid diameter from the equality ($v_{sed} = \xi v_{diff}$) in eq 4 as:

$$a_T = \frac{12\xi kT}{\pi d_c^3 (\Delta\rho) L_c} \qquad (5)$$

where L_c is a characteristic length scale of the diffusion process. In a porous medium, we can take this length scale as the average pore size, which we approximate here by the average grain size. Eq 5 allows us to predict the threshold centrifugal acceleration at which the transport behavior of colloids will be altered compared to normal gravity conditions. If the acceleration exceeds the one predicted by eq 5, we expect colloid transport to be affected by centrifugation.

Objective 2: *Evaluate the correction factor in the theoretical relationship for critical acceleration by using geocentrifuges*

The theoretical predictions were tested with colloid transport experiments conducted in water saturated columns. A geocentrifuge was used to vary the centrifugal acceleration and to determine the threshold acceleration beyond which colloid deposition was altered. We conducted a series of colloid filtration experiments using an acrylic column of 1.5-cm ID and 6.4-cm length. The entire column set up was placed on the platform of a 2-m geocentrifuge (Model C61-3 Civil Engineering Centrifuge, Actidyn Systemes, France) at the geocentrifuge laboratory of the Idaho National Laboratory, Idaho Falls, ID. We first conducted the experiments at 1 *g* (normal gravity) without spinning the centrifuge. In subsequent experiment conducted on the 2-m geocentrifuge, once the centrifuge reached its target centrifugal acceleration, the column influent was switched from a background solution containing no colloids to a solution that contained the colloids. The outflow solutions were collected using fraction collector and their concentrations were measured at different wavelengths for different colloids using a spectrophotometer (USB4000 spectrometer, Ocean Optics Inc.). The colloid breakthroughs were analyzed with the advection-dispersion model (ADE) including a first-order colloid deposition term. The statistical differences between estimated model parameters were evaluated with a one-way ANOVA and Tukey pairwise comparison using SAS.

PRINCIPAL FINDINGS AND SIGNIFICANCE

The experimental results and theoretical calculations indicate that colloid transport will be affected by altered force fields during centrifuge experiments. We expect colloid deposition to increase as acceleration exceeds a threshhold acceleration (because of increased sedimentation). This threshold acceleration is determined by the density difference between colloid and the fluid, the colloid size, and the pore size of the porous medium. Theoretical calculations illustrated that centrifuge colloid transport experiments in porous media are the most appropriate for small colloid specific density, colloid diameter, and pore diameters.

The colloids used in our experiments are representative for many subsurface colloids (bacteria, silicates and aluminosilicates, and (hydro)oxides). For inorganic subsurface colloids, such as silicates and iron oxides, colloid transport will be different in centrifuge experiments as compared to normal gravity conditions at already fairly low accelerations. In our experiments accelerations as low as 5 *g* changed the filtration behavior of colloidal hematite and silica. Because of their high particle density, (hydro)oxides will be most affected by centrifugal acceleration. Organic colloids, which have densities close to that of water, i.e., ≈ 1 to 1.4 g/cm^3, are less susceptible to sedimentation. However, natural organic colloids vary a lot in size. For instance, for viruses (particle density: 1.3 to 1.5 g/cm^3, diameter: 24 to 81 nm) threshold accelerations would be in the range of 200 to 10^6 *g*, for coarse and fine textures soils, respectively, so that most geocentrifuge experiments will not affect the transport behavior. Bacteria (particle density: 1.02 g/cm^3, diameter: 1 to 4 μm), however, because of their large size, have threshold accelerations between 1 and 250 *g*, for coarse and fine textures soils, respectively. Overall, the most sensitive parameter determining critical accelerations is the colloid diameter.

References

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