

Acceleration of Groundwater Remediation by Rapidly Pulsed Pumping: Laboratory Column Tests

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Abstract: Groundwater contamination is expensive to remediate, primarily because of the treatment time required. Increased treatment time can be caused by several contributing factors, such as sequestration in low-permeability regions of an aquifer, which can cause tailing and rebound phenomena. One such region consists of dead-end pores. Once a contaminant enters the dead-end pores, it remains in an eddy and only moves back into well-connected pores, from which it can be advected away, by molecular diffusion. A previous study with computational fluid dynamics suggested that rapidly pulsed flow demonstrated the potential to accelerate contaminant removal from dead-end pores. Here, we tested the recovery of a dissolved contaminant under steady and rapidly pulsed flow from a porous media column with crushed glass, which should have naturally formed dead-end pores. This test simulated the fluid dynamics in a flow constrained by a column without sorption; therefore, this tested the dynamics in the dead-end pores but did not include the complete dynamics of a remediation scheme. The results showed accelerated removal under rapidly pulsed flow, which is consistent with results from the computational model; however, there may be additional or alternative sources of the sequestered contaminant. DOI: [10.1061/\(ASCE\)EE.1943-7870.0001479](https://doi.org/10.1061/(ASCE)EE.1943-7870.0001479). © 2018 American Society of Civil Engineers.

Introduction

Groundwater remediation is a costly and time-consuming endeavor. The Environmental Protection Agency has estimated that \$209 billion is required over the next 30 years to address new sites (NRC 2013) in addition to the funds needed to continue and maintain old sites. Previously, the National Research Council had estimated the total monetary requirement for cleanup in the United States to be \$1 trillion (NRC 1994). In a survey of remediation sites, the median annual cost of operation was greater than one tenth of the capital cost (EPA 1999); since remediation times are on the scale of decades or more (NRC 1994), the total operational cost outweighs the capital cost, which makes time the dominant factor in overall cost. It is imperative for public health and water management that contaminated groundwater sites be remediated, and the most efficient way to expedite the process and decrease costs is through the development of techniques that decrease overall treatment time.

The most common remediation technique is pump and treat (P&T) (EPA 2007). A common problem in P&T and other techniques is tailing and rebound. When treatment begins, the contaminant concentration from the aquifer is high. Tailing is observed when the contaminant concentration of the effluent decreases faster than expected or modeled. Treatment is stopped at the end of the prescribed time or once the contaminant concentration in the effluent reaches the target, and the required contaminant is expected to have been removed. Rebound is observed when, after treatment has ended—often in regulatory checks—the tested effluent shows contamination levels higher than the target level (EPA 1996).

The sources of the newly emerged contaminants can be categorized as (EPA 1996; Harvey et al. 1994; Mackay et al. 2000; Mackay and Cherry 1989) nonaqueous phase liquids (NAPL) (e.g., Borden and Kao 1992), reactions (e.g., Colombani et al. 2009), sorption processes (e.g., Rabideau and Miller 1994; Saez and Harmon 2006), and matrix diffusion, which we examine in this work.

Matrix diffusion can describe any component of an aquifer with lower permeability, which includes volumes as small as specific pores that are poorly connected. Contaminants trapped in these regions can slowly diffuse back into the rest of the aquifer and cause a rebound in contaminant levels. In this paper, we specifically examined pores that are poorly connected to the main flow through the aquifer. We consider here two types of pores: those in which the flow enters the pore and flushes out all or most of the pore volume, and those in which the flow does not enter into the pore space. The former we term well-connected and the latter dead-end pores (Fig. 1). Contaminants sequestered in dead-end pores are a bottleneck to the remediation process.

These two types of pores were conceived to describe oil in porous media (Coats and Smith 1964), and later the system was extended to aquifers (van Genuchten and Wierenga 1976). The dynamics of these pores—specifically the rotational nature of dead-end pores—was studied by Kim et al. (2010). A similar system with trapped contaminants in dead-end pores is found in various manufacturing applications. Previous experiments using higher velocity flows (although still with a Reynolds number $R < 2,000$) to clean manufacturing surfaces resolved two mechanisms that were examined in this research (Fang et al. 1997; Nishimura et al. 1997).

Aref (1984) showed in an analytical solution of suitable two-dimensional, unsteady, laminar flow that there is chaotic motion termed chaotic advection. Pulsed flow, analyzed in the context of a numerical model of a dipole flow with Stokes flow, has shown chaotic motion of the diffusion of tracer dye (Jones and Aref 1988). This concept was used to show that pulsed flow can increase mixing and, therefore, groundwater remediation by bioremediation (Jones et al. 2002). Sposito (2006) studied chaotic advection in a vertical circulation well (a single well with two openings, one for outflow and one for inflow) and showed higher mixing efficiency.

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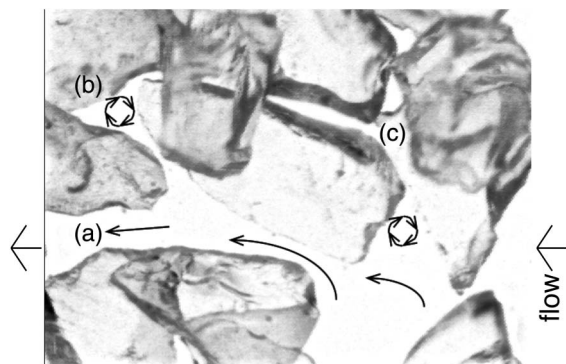


Fig. 1. Flow through porous media. The crushed glass media in cross-polarized light (image view: 2×10^{-3} m in width): (a) well-connected pores, which have a clear path through the space; (b) a dead-end pore, which does not have a flow through it but is adjacent to and influenced by the well-connected pore. The separatrix is the boundary between the well-connected pore and the water held in the dead-end pore; and (c) a dead-end pore configuration that is connected in the third dimension.

Zhang et al. (2009) also observed improved mixing in a laboratory-scale plume with spatially distributed wells.

The proposed rapidly pulsed pumping changes the flow dynamics within porous media. In a related technique, vibration of the soil matrix has been used to change flow dynamics. Biot (1956) developed the theoretical basis for vibration through a saturated matrix to change flow dynamics; however, this study did not consider any specific pore geometry such as the dead-end pore. Biot did highlight that, at low frequencies, fluids and solids can move in opposite directions. Reddi and Challa (1994) presented remediation experiments that focused on the removal of ganglia of NAPL in sand. The vibrations induced “cyclic pore pressures” that they hypothesized aided in the removal of the NAPL. They showed improved recovery under vibration compared with the nonvibrating control and found the recovery improved with increased amplitude. Roberts et al. (2001) tested vibrations of 25–100 Hz imposed on a solid matrix modeled by a column of sand initially filled with water. Trichloroethylene (TCE) was introduced to the flow as the tracer contaminant. The tests gave promising results for enhanced recovery; however, the authors stated that the “physical mechanisms responsible for the observed phenomenon are not fully understood.” Furthermore, in this process, the dead-end pores could not be fully considered because the pores were not fully/homogeneously contaminated with the technique of TCE introduction. Vogler and Chrysikopoulos (2002) performed experiments on solute transport in glass beads; they also introduced the solute tracer after the saturation of the medium, thus avoiding potential dead-end pores. In this pore geometry with introduced solute, the mobilization under vibration was higher than the stationary column. Lo et al. (2012) discussed vibrations in the context of NAPL at frequencies of 25–100 Hz and explained the pore volume fluctuations that could be responsible for enhanced movement of fluids. Their numerical and experimental results showed great promise in NAPL recovery.

The hypothesis tested here is that rapidly pulsed flow through an aquifer will accelerate the remediation of contaminants in P&T. This hypothesis has already been tested in an idealized system of one to three dead-end pores with computational fluid dynamics (CFD) (Kahler and Kabala 2016). Specifically, a steady flow through well-connected pores does not enter the dead-end pores but does induce a vortex within the dead-end pores. The separatrix, the boundary between the well-connected and dead-end pores (Fig. S1), is the surface between all of the streamlines in the

well-connected pores and all of the streamlines in the dead-end pores. A sudden increase in velocity causes the flow to push the separatrix further into the dead-end pores. When the flow sweeps deep into the dead-end pores it advects the sequestered contaminant to the well-connected pores in a process called a deep sweep. A sudden decrease in flow causes the vortex in the dead-end pores to emerge into the well-connected pores. The vortex circulates contaminants from the dead-end pores into a region that will soon be subject to the flow through the well-connected pores in a process called vortex ejection. The CFD models confirmed that rapidly pulsed flow recovers contaminants from dead-end pores faster than steady flow. These simulations were conducted at flow rates similar to what would be expected near pumping wells; however, further analysis showed that the phenomenon holds at velocities expected further from the wells. The work presented here expands on the theoretical and modeled work by extending the analysis to a collection of three-dimensional pore configurations through a physical model.

The fundamental mechanism improving P&T that was studied in this work was enhanced mixing between the well-connected pores and the dead-end pores. Enhanced mixing may also accelerate the performance of other remediation techniques such as bio-remediation. We hypothesized that rapidly pulsed pumping would cause deep sweeps and vortex ejections in a randomly arranged porous medium, which would increase the mixing between well-connected and dead-end pores and therefore accelerate groundwater remediation. This study tested the acceleration of the recovery of contaminants within a porous medium under rapidly pulsed flow. We specifically tested different types of theoretical soil grains: one that represents an unwashed medium, such as a glacial till, and one that represents a washed medium, such as riverine sediment.

Methods

We compared the action of the steady versus rapidly pulsed flow to remove contaminants from a column of an idealized porous medium. We constructed a column to simulate constrained flow (Fig. 2) with a 0.02-m (0.75-in.) inside diameter PVC pipe. The column was sealed at each end with a #60 brass screen to keep the particles contained but allow the flow through. While one end was fixed within an end cap and port to the downstream tubing, the top end was removable and held in place by a silicone sealant and a PVC screw-top fastener. The upstream tubing from the constant head tank to the column was 0.02-m (0.75-in.) inside diameter vinyl and the downstream tubing was 0.006-m (0.25-in.) inside diameter vinyl. The peristaltic pump was placed downstream and used latex tubing. Two types of media were tested: crushed glass and glass spheres. Crushed glass modeled an unwashed medium and glass beads modeled a washed medium. Glass was chosen because it is known to have low sorption with fluorescein dye (Kasnavia et al. 1999), which was the model contaminant, and therefore simulates fluid mechanics without sorption. This may be similar to deposits with low or no organic matter. Trials of steady and rapidly pulsed flow were alternated; any potential sorption to the plastic components should be distributed evenly between the types of trials. Both types were well sorted; the particles were sorted by standard soil sieves shaken on a geotechnical shaker for 20 min. The sorted particles were taken between sieve sizes 5.89×10^{-4} to 8.33×10^{-4} m (Numbers 30 and 20). Since the particles can fall past the larger sieve in such an orientation, the longest dimension of the particles may be larger than the larger sieve size; therefore, this particle size range was expected to form pores on the

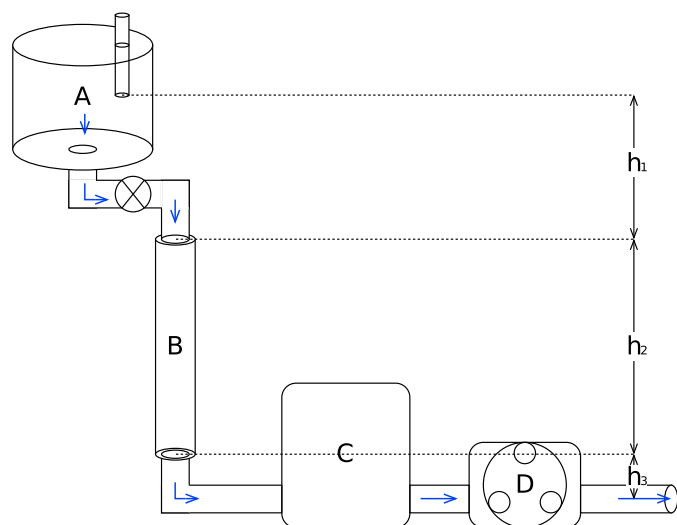


Fig. 2. Experimental setup (not to scale; from upstream to downstream) with a sealed tank (a) that has a static tap in the top that keeps the filled tank at a constant head of height $h_1 = 1$ m above the sample column. The vertical sample column (b) is filled with crushed glass or glass beads and has a cross-sectional area of $1.8 \times 10^{-4} \text{ m}^2$ and a length of $h_2 = 0.32$ m. The flow immediately enters the fluorometer (c) from the sample column; fluorescence, temperature, and calibrated concentration were recorded via an internal data logger. In pulsed flow trials, the next stage was the peristaltic pump (d) that induced the pulsed flow by pumping in the direction of flow; in steady flow trials, the peristaltic pump was not in the flow path. The free outlet was at approximately the same height as the instrumentation and pump, $h_3 = 0.1$ m below the outlet from the column. The arrows show the direction of flow through the apparatus.

order of 10^{-3} m, which was the same size as was examined in the CFD model. The porosities of the crushed glass and the glass beads were 0.455 and 0.400, respectively. The porous medium was removed, cleaned, and dried between each trial to removed residual model contaminant. The column was filled and manually compacted by striking the base against the bench until the column contained the desired solid particles. For each trial, the same mass of crushed glass or glass beads was used; however, the mass of crushed glass was not equal to the mass of the glass beads. Movement of the solid matrix was not expected because of compaction and the restriction of motion from the screens placed on both ends.

Fluorescein dye was used as the model contaminant (fluorescein sodium salt, Sigma-Aldrich, Milwaukee, Wisconsin; aqueous solution with municipal water). The model contaminant was prepared with fluorescein (powder) to a concentration of 10^{-4} kg/m^3 to ensure that the effluent was within the linear range of the fluorescent dye. Fluorescein simulated dissolved contaminants such as the soluble components of methyl tert-butyl ether (MTBE) or gasoline. Contaminant was introduced to our model porous media immediately prior to the experiment by flooding the clean, dry, porous medium with dye solution from the bottom of the column in order to fill all of the pore spaces and to minimize air trapped in the pores. Air was then removed from the other internal parts of the apparatus through purge valves. The test flow (either steady or rapidly pulsed) was then imposed on the sample. The outflow was analyzed with a fluorometer (Turner Designs 10AU field fluorometer, Sunnyvale, California) with a time resolution of one second. The 10AU measures fluorescence from the dye and temperature; the instrument

uses a calibration curve and automatic temperature correction to provide concentration. The instrument was calibrated according to manufacturer specifications by a two-point calibration with a blank and a known calibration standard within the linear range of fluorescence to concentration (0 – $10^{-4} \text{ kg m}^{-3}$ or 0.1 mg/L). The fraction of contaminant removed was computed by numerical integration of the concentration in the effluent and the volume flow rate, normalized by the original amount of contaminant that was added to the column.

The average flow velocity in the sample column was set to approximately 0.01 m s^{-1} in all trials in order to best replicate the flow rates expected in groundwater remediation in the vicinity of wells, match flow rates modeled in previous numerical work, and keep the flow away from the turbulent range for porous media; turbulence in some porous media can occur just over $R \approx 10$ (Masuoka and Takatsu 1996). The steady flow rate was adjusted with the valve just downstream of the constant head tank and above the tube to the porous medium column; this valve was used to reduce the introduction of additional mixing in the column. Pulsed flow rates were adjusted by means of the peristaltic pump (Manostat VERA Varistaltic Pump, Barrington, Illinois)—placed in line after the fluorometer—that generated a square wave pulse. The flow rate was also the minimum rate possible with the peristaltic pump. Based on the average cross-sectional area of the packed crushed glass or glass beads subtracted from the sample column, the trial flows corresponded to $R \approx 4.8$ – 6.7 . The peristaltic pump produced a pulse with a frequency of approximately 30 cycles per pore volume and a duty cycle of 0.12 (the ratio of the time at increased flow divided by the wave period; e.g., a square wave of amplitude a , a frequency of 1 Hz, and a duty cycle of 0.5 will spend 0.5 s at $+a$ and 0.5 s at $-a$), which was determined by a comparison of the output of the pump under pressure at the set rate. Unfortunately, the peristaltic pump did not allow for different frequencies of amplitude.

Results and Discussion

The column tests of steady versus rapidly pulsed flow were conducted through the crushed glass and glass beads with substantially different results. Three trials were performed with each flow type for each medium. More contaminant was removed from the crushed glass medium under rapidly pulsed flow than under steady flow. The average fraction of contaminant removed is plotted versus nondimensional time, expressed in pore volumes pumped, $\tau = t Q / v_{\text{pore}}$, where t is time, Q is the volume flow rate, and v_{pore} is the pore volume of the column, for crushed glass (Fig. 3). The results show that pulsed flow recovered 100% of the fluorescein and that the steady flow recovered 89% of the fluorescein after about 10 pore volumes pumped. The 30 cycles per pore volume, in this apparatus, equated to roughly 6 Hz, which was far below the acoustic frequencies over 60 Hz previously tested (Biot 1956; Vogler and Chrysikopoulos 2002).

Steady flow continued to recover more contaminant as the experiment progressed; therefore, the biggest benefit that rapidly pulsed flow offers is in the rate of recovery. The rapidly pulsed flow recovered all of the detectible contaminant by the time that steady flow recovered almost 88% of the contaminant. To better illustrate this recovery, we defined the speedup factor to be the ratio of the time that it took for steady flow to achieve a given fraction of contaminant removed to the time that it took for rapidly pulsed flow to achieve the same fraction removed: $\tau_{\text{steady}}|_f / \tau_{\text{pulsed}}|_f$, and plot that versus the fraction removed (Fig. 4). In Figs. 3 and 4, it is evident that the average rapidly pulsed flow did not remove contaminant as

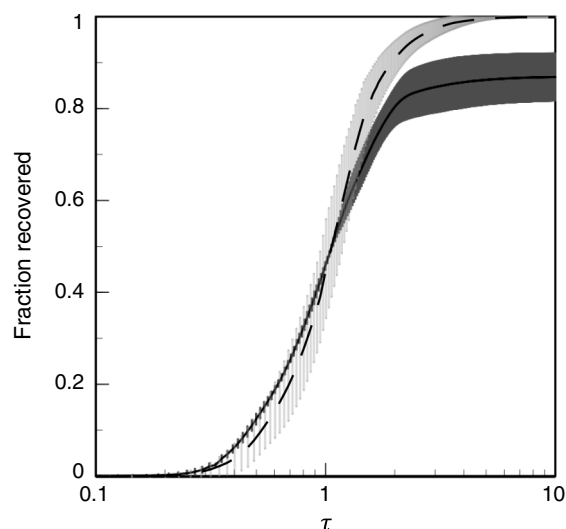


Fig. 3. Fraction of the original contaminant removed from the column of crushed glass under steady (solid) and rapidly pulsed (dashed) flow. The standard deviation is shown by error bars.

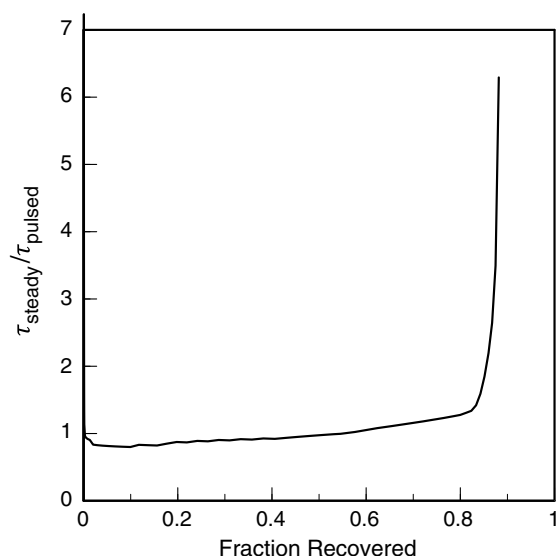


Fig. 4. Speedup factor, defined as the time of remediation under steady flow divided by the time of remediation under rapidly pulsed flow for a given fraction of contaminant removed. The speedup factor shows how many times faster the rapidly pulsed flow would achieve a given fraction recovered than the steady flow.

quickly as steady flow from the column of crushed glass at the start of the trials. In two of the three trials, recovery from the column was faster under steady flow for approximately the first 40–80% of the contaminant (or, roughly the first two pore volumes), which is evidenced in the average of the trials in Fig. 3. The column chamber had significant edge effects; there was an artificially well-connected channel adjacent to the edge. In the well-connected region, there was no benefit to the rapidly pulsed flow, and it is possible that much of the contaminant advected through the column. These flow rates best simulate the flow near injection or extraction wells. Lower flow rates have been explored numerically (Kahler and Kabala 2016) and showed that dead-end pores are present at flows with Reynolds numbers as low as $R = 10^{-2}$, and they shrink by

less than 10% compared to the Reynolds number of the flows considered in these experiments.

Unlike the crushed glass, the recovery of contaminant from glass beads under pulsed flow was not significantly faster and did not recover more contaminant than steady flow. The rapidly pulsed flow performed similarly to steady flow for the glass beads (Fig. S2). One possible explanation for this difference between the media is that, with the glass beads, the pore shape is curved and does not contain abrupt edges like the crushed glass. This is consistent with CFD models of contaminant recovery from pores formed by modeled glass beads (Fig. S1) that showed there was no improvement with rapidly pulsed flow versus steady flow (Fig. S3) (Kahler 2011). The slightly faster recovery shown in the average of three trials may have been due to complex dead-end pore formations or within the natural variation of the column tests.

As an illustration of one of the mechanisms that is responsible for accelerated recovery in an unwashed medium, a pore system was constructed that consisted of a well-connected pore modeled as a channel and two dead-end pores modeled as two departures from the main channel. The two-dimensional model was initially covered with a thin layer of blue dye. An oscillatory flow was passed through the well-connected pore, and enhanced contaminant mixing by the deep sweep and vortex ejections (Kahler and Kabala 2016) were observed. The model does not attempt to reach complete similitude with that of a natural aquifer; however, it does illustrate the mechanisms expected in the crushed glass (Fig. S4 and Movie S1).

Conclusions

Traditional pump-and-treat remediation uses steady flow rates, or rates adjusted on a scale on the order of months or years, to recover contaminants from aquifers. Contaminants sequestered in dead-end pores can only escape by molecular diffusion across the separatrix and out of the dead-end pores. Porous medium column experiments were conducted with fluorescein dye as a practically nonsorbing conservative tracer and glass to model the solid matrix. Crushed glass modeled unwashed media such as glacial deposits and fractured rock. Glass spheres modeled washed media such as fluvial deposits. Rapidly pulsed flow through a porous medium column filled with crushed glass was shown to recover more contaminant in the trial period and was able to reach higher removal levels faster than steady flow. Roughly 11% more contaminant was recovered with the rapidly pulsed pumping, and to remove more than 89% of the contaminant from the column, rapidly pulsed flow was at least seven times faster. These improvements were not seen with the glass spheres. The improvement in crushed glass and lack of improvement in glass beads was consistent with previous numerical studies on these two geometries. By this comparison, we hypothesize that the deep sweep and vortex ejections are responsible for the observed improved contaminant recovery. In actual P&T configurations, extraction and injection wells are used in an aquifer, which result in a significantly different flow regime than was examined in this experiment. Aquifers contain different and varied pore geometries and heterogeneous soil. Further experimentation is required to test rapidly pulsed pumping in complex pore structures. Furthermore, different contaminants, such as those with sorption and nonaqueous phase liquids need to be included. Realistic pumps should be examined for delivery of rapidly pulsed flow and applied to aquifers. These results have significant implications for shortening the duration of treatment and improving the amount of contaminants removed by pump-and-treat remediation.

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Supplemental Data

Figs. S1–S4 and Movie S1 are available online in the ASCE Library (www.ascelibrary.org).

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