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Development of Radon-222 as a Natural Tracer for Monitoring the Remediation of NAPL Contamination in the Subsurface

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Research Objective

The objective of this research is to develop a unique method of using naturally occurring radon-222 as a tracer for locating and quantitatively describing the presence of subsurface NAPL contamination. The research will evaluate using radon as an inexpensive, yet highly accurate, means of detecting NAPL contamination and assessing the effectiveness of NAPL remediation. Laboratory, field, and modeling studies are being performed to evaluate this technique, and to develop methods for its successful implementation in practice.

Research Progress and Implications

This report summarizes work that has been accomplished after 1-year of a 3-year project. The research to date has included radon tracer tests in physical aquifer models (PAMs) and field studies at Site 300 of the Lawrence Livermore National Laboratory, CA, and Site 100D at Hanford DOE Facility, WA. The PAM tests have evaluated the ability of radon as a tracer to monitor the remediation of TCE NAPL contamination using surfactant treatment, and oxidation with permanganate. The surfactant tests were performed in collaboration with Dr. Jack Istok and Dr. Jennifer Field and their EMSP project "In-situ, Field-Scale Evaluation of Surfactant Enhanced DNAPL Recovery Using a Single-Well-Push-Pull Test." This collaboration enabled the EMSP radon project to make rapid progress. The PAM surfactant tests were performed in a radial flow geometry to simulate the push-pull-method that is being developed for surfactant field tests. The radon tests were easily incorporated into these experiments, since they simply rely on measuring the natural radon present in the subsurface fluids. Two types of radon tests were performed: 1) static tests where radon was permitted to build-up to steady-state concentrations in the pore fluids and the groundwater concentrations were monitored, and 2) dynamic tests where the radon response during push-pull surfactant tests was measured. Both methods were found to be useful in determining how NAPL remediation was progressing.

Methods for conducting the surfactant test in the PAMs are described in the Progress Report of Istok and Field. The PAMs were packed with subsurface solids from Site 300, which emanated enough radon for its use as a natural tracer. Radon concentrations in 5 ml groundwater samples from the PAM were accurately measured using a low background Liquid Scintillation Method with Alpha-Beta-Discrimination Counting. Studies were performed with no NAPL present and with the addition of 5% TCE saturation to a portion of the PAM. Radon concentrations under static (no flow conditions) were a factor of two lower in the region that contained NAPL. The reduction in radon concentration, however, was not as low as predicted by partitioning theory. These results were consistent with TCE measurements and indicate that the TCE contamination was not uniformly distributed in the PAM. Results of dynamic tests showed radon transport being retarded due to the partitioning into the NAPL TCE, consistent with transport theory. Both static and dynamic radon tests were performed during the course of the push-pull surfactant experiments. The remediation with DPDS surfactant resulted in little change in the radon concentration distribution in the PAM, indicating minor amounts of NAPL TCE were removed. This result is consistent with direct TCE mass balances that were performed.

PAM tests were also performed of TCE NAPL remediation using permanganate oxidation. Experiments were conducted using a push-pull test methodology for the addition of the permanganate, similar to that used for the surfactant studies. Low and high concentrations of permanganate were injected during the course of the study. Static radon tests showed radon concentrations, that were initially reduced in the NAPL contaminated region of the PAM, increasing with permanganate additions. The most dramatic increases occurred near the end of the PAM where the permanganate was injected. The results agree with chloride mass balances that indicated oxidation of 35% of the in-place TCE. Dynamic push-pull tests, also show radon concentration responses changing during the course of the remediation. Water lacking radon was injecting during the push phase of the push-pull tests. During the pull-back phase, radon concentrations increased more rapidly as NAPL remediation progressed, consistent with transport theory. The results from the surfactant and the permanganate PAM studies illustrate the potential for using radon as a natural tracer to monitoring the progress of NAPL remediation in the field.

Field studies at the LLNL Site 300 included three preliminary radon tracer push-pull-well tests at the site. Preliminary groundwater radon surveys conducted prior to the start of the EMSP project showed depressed radon concentrations in region of the aquifer where NAPL contamination is expected. Tests were performed in wells containing low, intermediate, and high radon concentrations by injecting radon free water into the wells along with a bromide tracer. Consistent with transport theory, the radon concentrations increased most rapidly in the well with high radon concentration groundwater, and remained essentially constant in the well with low radon concentration groundwater. The results indicate the presence of NAPL in the region with low radon concentrations. The results illustrate how the radon method can be successfully employed with other push-pull tests for remediation that are being developed.

Studies at the Hanford Site were performed as part of test using partitioning gas tracers, to determine if trapped gas is present in the aquifer at Site 100D. These tests provided an opportunity to study a radon partitioning process that is similar to that which occurs with NAPLs. Two tests were performed: 1) a push-pull-single well test, and 2) a radial injection transport test with sampling at a multi-level monitoring well. Radon free groundwater was injected in both tests. Again the radon method was easily incorporated into these tests and required only the monitoring of radon concentrations and no addition of tracer. The radon response indicated the presence of trapped gas in the aquifer, consistent with results obtained with partitioning tracers that had to be carefully added, such as SF₆.

Planned Activities

Radon studies will be continued using the PAMs for NAPL remediation using push-pull methods. A new set of surfactant studies will be performed using Aerosol MA, which promises to be a more effective surfactant for TCE NAPL remediation. Radon tests will also be continued in PAM remediation experiments via permanganate oxidation of TCE. We will also initiate studies in a 2-D horizontal physical model that is packed with aquifer solids from the Hanford Site. The model is representative of aquifer conditions where regional flow is induced. Distributed TCE NAPL zones will be created in the 2-D model, and the groundwater radon concentration distribution will be determined before and after TCE is injected, and after TCE is remediated using permanganate and/or Aerosol MA. Push-pull tests will be performed within the 2-D model to simulate field scale tests.

We also plan to conduct experiments with Dr. Mart Oostrom at the PNL Laboratory at Hanford. Dr. Oostrom has a two dimensional vertical model with a Dual Energy Gamma System for measuring TCE and water saturations, permitting direct comparisons with the radon method for estimating TCE saturations. A radon survey is also planned for Savannah River, in collaboration with Joseph Rossabi, Westinghouse.

Other Access To Information

Semprini, L.M. Cantaloub, S. Gottipati, O. Hopkins, and J. Istok. Radon-222 as a Natural Tracer for Quantifying and Monitoring NAPL Remediation. Proceedings: First International Conference on the Remediation of Chlorinated and Recalcitrant Compounds, May 18-21, 1998, Monterey, CA (In Press).