Project ID: **60158**

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

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Development of Radon 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 for using naturally occurring radon-222 as an inexpensive partitioning tracer for locating and quantitatifying nonaqueous phase liquid (NAPL) contamination in the subsurface, 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 1/2 years 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, Site 100D at Hanford DOE Facility, WA, The Dover National Test Site, DE, and the Weyerhauser-Sycan Site, Beatty, OR.

During the past year PAM tests were completed that evaluated the ability of radon as a tracer to monitor TCE NAPL remediation by permanganate oxidation. The tests were performed in radial flow geometry to simulate the flow field during field push-pull tests used to evaluate the permanganate oxidation technology. The radon tests were easily incorporated into these experiments, since they simply rely on measuring the natural radon present in the pore fluid. Two types of radon tests were performed: 1) static tests, where radon was permitted to build-up to steady-state concentrations in the pore fluid before being measured, and 2) dynamic push-pull-tests, where the radon concentration response was monitored by injecting water that lacks radon into the PAM and pulling the water back and measuring radon concentration as a function of volume of water extracted. Both methods were found to be useful in monitoring the progress of NAPL remediation. The PAM was packed with subsurface solids from LLNL 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 then with the addition of 5 vol % TCE NAPL to a portion of the PAM. Repeated push-pull additions of high and low concentrations of permanganate were injected into the PAM during the course of the study and both static and dynamic radon tests were performed. The results of the static profiles are presented in Figure 1. Radon concentrations under static (no flow) conditions after TCE addition, but prior to

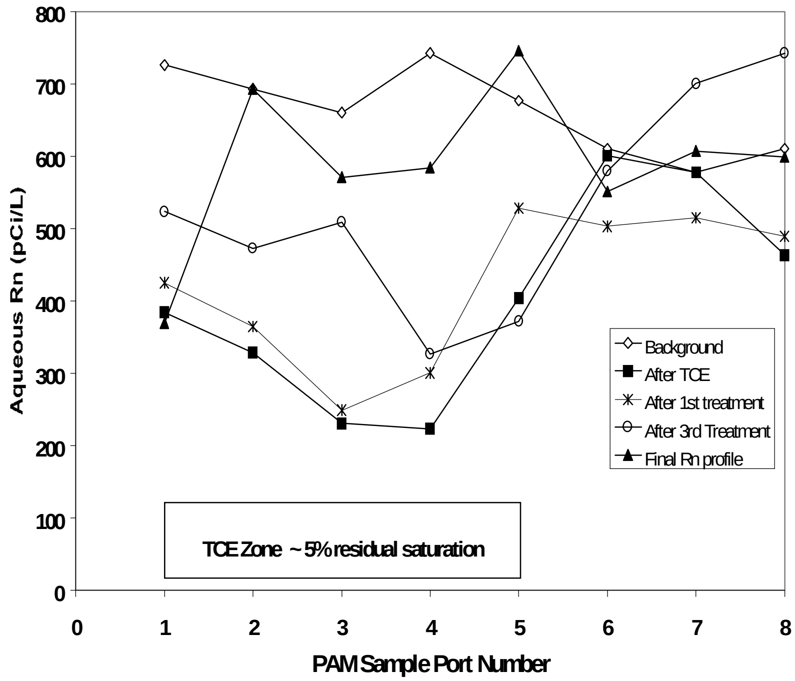

Figure 1. Static profiles of radon concentration in a PAM containing NAPL TCE that was remediated with successive addition of permanganate.

permanganate treatment were a factor of two to three lower in the region that contained TCE. The reduction in radon concentration, however, was not as low is predicted by partitioning theory. Radon concentrations increased with successive injections of permanganate, and eventually reached levels that existed before TCE addition. The reason for the low radon level at sampling Port 1 is not known, but it may be associated with a build-up of carbon dioxide gas in this narrow region of the radial PAM. If a trapped gas phase is present, radon would partition into the gas phase and aqueous radon concentrations would be reduced. Overall the radon results indicated effective permaganate oxidation of the NAPL TCE. The results are consistent with observations of permanganate utilization, which show extensive utilization when radon deficits were observed, and little permanganate utilization after the radon reached background levels.

Dynamic push-pull tests also show radon concentration responses changing as remediation proceeds. During the push (injection) phase water lacking radon was injected to probe the region that contained NAPL; during the injection phase flow was reversed and radon concentrations were measured in the extracted water. The results of these push-pull tests are presented in Figure 2.

The radon concentrations increased with successive treatments of permanganate, especially when the ratio of the extracted volume to injected volume was greater than 1.0. The results are consistent with those expected during the course of NAPL remediation. Consistent changes in the breakthrough patterns were not observed during these tests. This may result from dynamic changes in flow patterns, which were also indicated by concentrations of the bromide tracer. The results, however, do indicate that push-pull

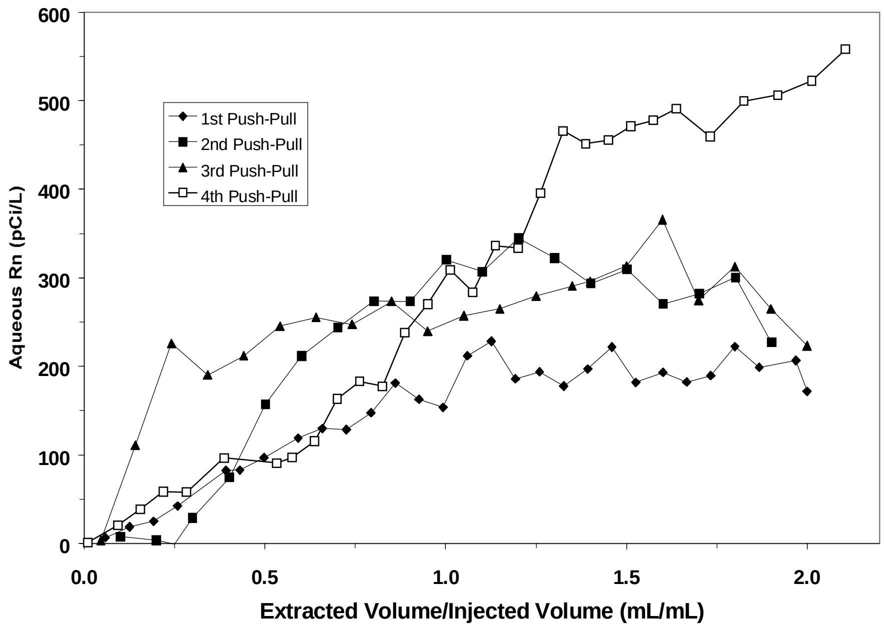

Figure 2. Radon concentrations as a function of the ratio of extracted to injected volume of a push-pull test.

tracer tests utilizing radon have the potential for monitoring the progress of NAPL remediation. The results of these tests would indicate that the amount of fluid injected and extracted is an important consideration when using this method.

Currently we are determining the TCE remaining after treatment by measuring the TCE content in core samples. We are also measuring radon emanation from core samples to determine if changes occurred as a result of the remediation process.

Studies in a 2-D horizontal PAM (2 m x 4 m x 0.2 m) that is packed with aquifer solids from the Hanford Site were initiated. The PAM model is representative of aquifer conditions where regional flow can be induced. Initial static radon concentrations through the PAM have been determined and push-pull tests in wells surrounding the zone where NAPL is to be added have been completed. The results from the push-pull tests show very reproducible radon and bromide responses, with radon being transported similarly to the non-partitioning bromide tracer, as is expected with no NAPL present. Distributed TCE NAPL zones will now be created in the 2-D model and the static radon concentration distribution will be determined and push-pull tests will be repeated. TCE NAPL contamination will then be remediated using permanganate and the static and dynamic radon tests will be performed.

A field evaluation of the push-pull radon method was performed at the Weyerhaeuser-Sycan Site in Beatty, OR. Perchloroethene (PCE) NAPL contamination was suspected at a source zone of contamination. Push-pull studies were conducted along with permanganate injection experiments. Direct analysis of core samples for NAPL contamination was also performed. The push-pull tests showed similar radon and bromide breakthrough responses, indicating that NAPL was not present in the source zone. Little permanganate was utilized in the test zone, and core samples analysis show no detectable NAPL contamination, which are both consistent with the radon results.

A radon field survey was also performed of water from Test Cell 3 at the Dover National Test, DE. The test cell had been purposely contaminated with PCE. The radon survey was conducted under static flow conditions in the cell after no flow had occurred for a period of one month. Obtaining good groundwater samples from the test cell proved to be difficult due to plugging of sample points within the test cell. Many samples appeared to be stripped of both PCE and radon. In several location samples were obtained that had low radon values and high PCE concentrations, which is expected if a NAPL zone was present. However, due to sampling problems that were encountered it is difficult to draw conclusions from these data. Over the past several months a study was performed of NAPL remediation using alcohol flushing methods. We are currently planning to monitor the radon concentrations in the cell to determine if changes occurred as a result of the flushing experiments.

PLANNED ACTIVITIES: Studies in the 2-D horizontal PAM model will evaluate the radon response to NAPL remediation with permanganate. We also plan to construct a vertical PAM and perform radon studies in a layered system. We will investigate an industrial site that has NAPL contamination, using push-pull tests. We also will continue to investigate Test Cell 3, at the Dover National Test Site. Successive NAPL addition and remediation methods are planned in the test cell.

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