

Research Objectives

The primary purpose of this project is to apply the use of isotopic measurements to develop and test conceptual models of the migration of radionuclides through the unsaturated zone and into groundwater. These models form the basis for making assessments of the environmental impacts of the contamination on short and long time scales, and for assessing how useful a particular remediation strategy will be. Specifically, we wish to determine whether:

1. The isotopic compositions of O, H, C, and Sr in near-field groundwater samples can be used to evaluate the presence of- and sources of waste fluids, and therefore evaluate the fluid transport independent of the contaminants.
2. The isotopic compositions of O, H, C, Sr, N and U in near-field groundwater samples can be used to determine whether chemical processes (dissolution-precipitation, evaporation, microbial processes) significantly perturb transport of radionuclides.

The Hanford site was chosen for this work because it is probably the best site in the DOE complex to allow us to understand the behavior of the natural isotopic tracers. A very large amount of water containing radioactive and other contaminants was discharged to the vadose zone at Hanford starting in the 1940's and continuing until the 1980's. Groundwater plumes of tritium, technetium, iodine-129, nitrate and other species confirm that much of the released water reached the water table. These contaminants threaten the nearby Columbia River.

Research Progress and Implications

This report summarizes the results of work for this project through the first 8 months of FY2001 (the first year of a 3-year project). This is a renewal project (the initial project was EMSP #55351) and builds on preliminary findings from that research. Most of the work for the initial project was conducted at the Idaho National Engineering and Environmental Laboratory, but during the final year (FY1999) we received greater than 200 groundwater samples from the Hanford site.

Finishing work on the FY1999 Hanford sample and implementing a new sampling and analysis plan has been the primary focus of our efforts during this fiscal year. In summary:

1. FY1999 Samples
 - a. Completion of analyses: Analyses of the strontium and oxygen isotope ratios of all of the FY1999 were completed. In addition, analyses were done of the hydrogen isotope analyses of a subset of the samples.
 - b. Compilation of results: The isotopic analyses of all FY1999 samples were compiled into tables and plotted on figures for interpretation and planning of new sampling efforts.
 - c. Sample disposal: All residual samples from the FY1999 sampling program have been disposed of in accordance with ES&H requirements.
2. Presentation of Results
 - a. A summary of the results available from the FY1999 sampling was presented at the FY2001 EMSP Vadose Zone Principal Investigator Workshop at PNNL in November.
 - b. An interim report containing the results of the initial EMSP project was submitted in April.
3. FY2001 Sampling and Analyses
 - a. New sampling plan: A plan for new samples to be collected during this fiscal year was implemented (including setting up a subcontract to cover costs at PNNL). To date, 95 samples have been received at LBNL. Approximately 50 additional samples are expected before the end of the fiscal year.

- b. Analyses: Isotopic analyses of the new FY2001 samples are progressing. The carbon isotope ratios of dissolved inorganic carbon have been completed for all except the latest sample shipment. Hydrogen and oxygen isotope measurements of the water are underway (~50% complete) and strontium isotope measurements have begun. Uranium isotope measurements will begin soon (the analytical technique is currently being developed).

So far, the most interesting implication of our results has been the discovery that the strontium isotope ratios of the groundwater represent a sensitive indicator of the source and infiltration pathway of the water. The groundwater at the site has distinctly different $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (~0.707) than the Columbia River water (~0.714) that was used for work at the site. In addition, the sediments in the vadose zone at Hanford have strontium isotope ratios greater than or equal to the river (up to 0.721). As a result, in areas where significant amounts of process water were released at the surface, the strontium isotope ratios of the groundwater are shifted towards higher values. One of our goals is to be able to use this data to determine the proportion of process water in the groundwater and the pathway it used to travel through the vadose zone. Other isotopic measurements that we have made (e.g., hydrogen and oxygen) add to this understanding. In addition, we anticipate that uranium isotope measurements will significantly enhance the conclusions we can make.

Planned Activities

During the next fiscal year, we plan to focus additional groundwater sampling in the vicinity of the 200W and 200E tank farms. The purpose of this is to determine whether we can detect groundwater impacts from leaks from specific sources (e.g., tanks, infiltration ponds). We also plan to begin regular sampling of groundwater in the vicinity of the SLADS site (the active disposal site for water with low-level of tritium contamination). This will allow us to determine the behavior of other natural isotopic tracers (e.g., C, Sr, U) in relationship to the tritium (a conservative tracer) that is being released.

We also intend to expand our analytical efforts. We plan to begin analyzing the isotopic compositions of nitrate to use as a tracer of different waste streams and a potential indicator of subsurface biological activity. In addition, we would like to begin analyzing the isotope ratios of other radionuclides that could be potential tracers for different processes. This will involve development of the techniques for the multi-collector ICPMS.

Information Access

Conrad, M.E. and DePaolo, D.J., 2000, Isotopic Tracers for Waste Fluid Tracking and Fluid-Soil Interaction: Hanford, Washington. Presented at FY2001 EMSP Vadose Zone Principal Investigator Workshop, <http://www.pnl.gov/emsp/presentations/conrad2/sld001.htm>