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Characterization of Contaminant Transport Using Naturally-Occurring U-Series Disequilibria

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Research Objective

The goal of the research is to study the migratory behavior of contaminants in subsurface fractured systems using naturally occurring uranium- and thorium-series radionuclides as tracers under in-situ physico-chemical and hydrogeologic conditions. Naturally occurring U- and Th-series disequilibria can provide information on the rates of adsorption-desorption and transport of contaminants as well as on fluid transport and rock dissolution in a natural setting. We are developing a realistic model of contaminant migration in the Snake River Plain Aquifer beneath the INEEL by evaluating the retardation processes involved in the rock/water interaction. The major tasks are to: 1) determine the natural distribution of U, Th, Pa and Ra isotopes in the groundwater as well as in rock minerals and sorbed phases, and 2) study rock/water interaction processes using U/Th series disequilibria and a statistical analysis-based model code for the calculation of in-situ retardation factors of radionuclides and rock/water interaction time scales. This study will also provide an improved understanding of the hydrogeologic features of the site and their impact on the migration of contaminants.

Research Progress and Implications

This report summarizes results after 20 months of a 36-month project. Studies performed at LANL include analysis of the long-lived nuclides ^{238}U, ^{235}U, ^{234}U, ^{230}Th, ^{226}Ra, ^{232}Th, and ^{231}Pa by thermal ionization mass spectrometry (TIMS). Studies performed at the University of Southern California include the measurement of short-lived naturally occurring radionuclides by decay-counting techniques and the development of models to predict the migration behavior of these radionuclides. Initial efforts began with analysis of 31, 0.5L water samples obtained through routine sampling by USGS and INEEL personnel. One significant observation from these data is that $^{234}U/^{238}U$ activity ratios are highest in waters that emanate from local recharge areas to the northwest of the INEEL and they decrease in the direction of groundwater flow. Contours of high $^{234}U/^{238}U$ ratios delineate preferential flow paths extending southward from the local recharge areas. Our uranium data have allowed delineation of these preferential flow pathways in greater detail than obtained by other methods. The contour map of $^{234}U/^{238}U$ ratios also identifies isolated pockets with low $^{234}U/^{238}U$ ratios. These are suspected to represent groundwater which has undergone more extensive water/rock interaction as a result of either longer residence times of faster reaction rates. These results are important for understanding the hydrologic context of the site and crucial for interpreting our complete data sets for the U and Th decay chains.

Because we found insufficient Th, Pa and Ra in the small samples obtained through routine USGS sampling, we had to modify our sampling strategy in order to obtain samples sufficiently large to allow measurement of the short- and long-lived nuclides of the U and Th decay chains. This effort was not anticipated in our original research plan, but we have successfully collected large-volume groundwater samples (~1000 liters) from 12 wells from the site. Radioisotopes of thorium (^{232}Th, ^{230}Th, ^{228}Th, and ^{234}Th), radium (^{226}Ra, ^{228}Ra, and ^{224}Ra) and polonium (^{210}Po) were pre-concentrated in the field by passing the samples through two cartridge filtration units (connected in series at each well) containing MnO_2-impregnated acrylic fiber adsorbers. An additional 12L of water was collected for Th, Pa and Ra concentration measurements by TIMS. We have now obtained a full suite of U- and Th-series nuclides from these large-volume samples. Isotope ratio plots show some clear trends. In general, the degree of disequilibria among the U, Th and Ra isotopes decreases along flow pathways

and away from local recharge drainages to the northwest of the site. The short half-lives of some of these nuclides (e.g., ^{224}Ra $t_{1/2}$ = 3.66 days) indicates that adsorption/desorption must be important in controlling some of the isotope ratios. Trends among more conservative, long-lived species (e.g., U) are taken to reflect mixing and allow estimation of relative volumes of different groundwaters.

We have successfully estimated the degree of retardation due to rock-water interaction in the groundwater system at INEEL by modeling the observed radioactive disequilibria. Our model results indicate that adsorption of radium by the aquifer solids at the site takes place on time scales of a few tenths to several seconds, about 1-2 orders less than those of thorium. The desorption times, on the other hand, are several days for radium and several years for thorium. Based on these sorption-desorption rate characteristics, we estimate that retardation factors (i.e., the ratio of flow rate of fluid to those of dissolved radionuclides) are of the orders of 106 for ^{230}Th, 104 for ^{226}Ra, and 103 for ^{238}U. The model predicts that radionuclides sorbed on solids, which are exchangeable with the dissolved species through sorption/desorption, play an important role in affecting the decay-series disequilibrium in groundwater systems. The model also indicates that the major source of ^{222}Rn in groundwater from the study area originates from decay of ^{226}Ra sorbed on aquifer solid surface, rather than the previously held notion of direct alpha recoil from the solid ^{226}Ra pool.

The retardation-factor estimates that we have derived contribute to our understanding of the disposal and migratory behavior of radioactive wastes at INEEL. These estimates allow us to delineate a rather large range of migration rates for possible contaminants in groundwater at the site and suggest that three groups of wastes can be identified in terms of their mobility. The migration of radionuclides with high ionic potential (readily hydrolyzed) such as thorium and lanthanide fission products is heavily retarded. The degree of retardation for alkali- and earth alkali-radionuclides (e.g., ^{133}Ba, ^{90}Sr,and ^{137}Cs), is such that these nuclides are unlikely to be transported far away from the contaminated areas. Uranium (and perhaps many transuranic radionuclides) displays relatively high mobility in the aquifers we have studied.

Planned Activities

In the coming months we plan to analyze ^{210}Pb in the samples at hand. Plans are being formulated to carry out additional large-volume groundwater sampling from other wells at the site, and to collect samples of ~5-liter size for the measurement of ^{222}Rn in order to further constrain the alpha-recoil input of radionuclides to the groundwater medium. Upon completion of the fluid analyses, we also intend to obtain information on the activities of U, Th, Ra and Pb in rock samples from drill cores retrieved by the USGS - information that is germane to our modeling the transport in the study area. Fourteen samples from two cores have been collected and we plan to sample a third. The model code for contaminant transport will be further developed using these new data.

Other Access To Information

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