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Uranium-Series Constraints on Radionuclide Transport and Groundwater Flow at the Nopal I Uranium Deposit, Sierra Peña Blanca, Mexico

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Abstract – Uranium-series data for groundwater samples from the vicinity of the Nopal I uranium ore deposit are used to place constraints on radionuclide transport and hydrologic processes at this site, and also, by analogy, at Yucca Mountain. Decreasing uranium concentrations for wells drilled in 2003 suggest that groundwater flow rates are low (< 10 m/yr). Field tests, well productivity, and uranium isotopic constraints also suggest that groundwater flow and mixing is limited at this site. The uranium isotopic systematics for water collected in the mine adit are consistent with longer rock-water interaction times and higher uranium dissolution rates at the front of the adit where the deposit is located. Short-lived nuclide data for groundwater wells are used to calculate retardation factors that are on the order of 1,000 for radium and 10,000 to 10,000,000 for lead and polonium. Radium has enhanced mobility in adit water and fractures near the deposit.

I. INTRODUCTION

Geological barriers are an important possible mechanism for the isolation of radioactive contaminants from the biosphere. The effectiveness of such barriers can be characterized by measurements of retardation factors, e.g., the transport rate of contaminants relative to that of groundwater. As part of the OSTI Natural Barriers program of the U. S. Department of Energy, we are performing this characterization using naturally-occurring decay-series radioisotopes as analogs for radioactive contaminants and an abandoned uranium mine as an analog to the proposed high-level nuclear-waste repository at Yucca Mountain, Nevada. Our study area is the Nopal I uranium deposit in Chihuahua, Mexico, which is an excellent analog for evaluating the fate of spent fuel and its associated actinides and fission products over long time scales for the proposed repository. Uranium-series nuclide abundances in groundwater samples can be used to constrain recent radionuclide transport and groundwater hydrologic characteristics/processes at this site, and, by analogy, at Yucca Mountain.

II. WORK DESCRIPTION

The general location of the uranium ore deposit, along with saturated zone water sample locations is shown in Figure 1. The aquifer rocks consist of the Pozos conglomerate for most of the wells, with Cretaceous limestone for PB4 and Pozos Ranch. Unsaturated zone (UZ) water sample locations from the mine adit are shown in Figure 2. The mine adit is located at the +00 level of the deposit near the location for PB1. The UZ rocks above the mine adit consist of welded tuffs that host the ore deposit. Groundwater samples were generally collected intermittently during 2000-2005, whereas groundwater samples from the wells drilled in 2003 (PB1,

PB2, PB3) proximate to the deposit have been collected periodically from 2003-2005.

Saturated zone water samples were collected by bailer or pump. Unsaturated zone water samples were collected from the mine adit via a drip collection system consisting of plastic sheeting which was designed to funnel water into plastic bottles. Water samples for uranium concentration and ²³⁴U/²³⁸U analyses were filtered (0.20 μm or 0.45 μm filters) soon after collection and acidified to a pH of 1-2 with high purity nitric acid for long-term storage. For analyses of short-lived uranium-series nuclides, in April 2005 we sampled large-volumes (>1000 liters) of groundwater from three wells (Peña Blanca well, Pozos Ranch well, and PB4, respectively) near the Nopal I uranium deposit site, using an in-situ Mn-cartridge filtration technique which effectively adsorbs these nuclides [1]. Groundwater samples were also collected from these wells in a sealed 1L glass bottle for analysis of ²²²Rn by α-scintillation counting [1].

Manganese oxide and adsorbed radionuclides in Mn-cartridges were dissolved with 4 M HCl solution in the presence of ²²⁹Th and ²⁰⁹Po as yield tracers of Th and Po isotopes, Ba²⁺ and Pb²⁺ as yield tracers of Ra and Pb isotopes, and NH₂OH·HCl as a reducing agent. Thorium, Po, and Pb were separated from Ra and Ba by coprecipitation with Fe(OH)₃. Polonium was autoplated onto a silver planchet and counted for ²¹⁰Po by α-spectrometry using a surface-barrier Si detector. After Po autoplating, the sample solution containing Th and Pb was counted for ²³⁴Th and ²¹⁰Pb by γ-spectrometry using a high-resolution, well-type intrinsic Ge detector. Thorium was then further separated and purified by anion exchange for α-spectrometric measurements of the α-emitting Th isotopes (²³²Th, ²³⁰Th, and ²²⁸Th), which are still in

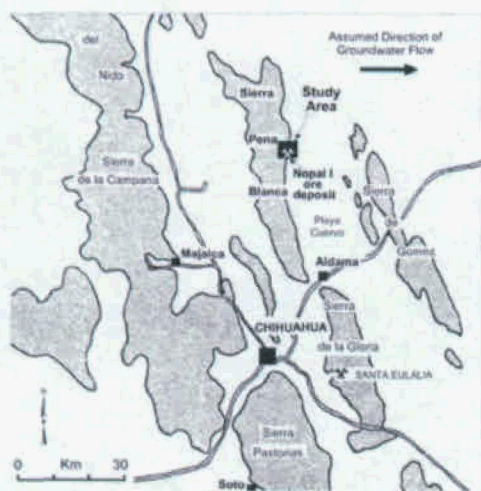


Fig. 1. General location of study area and locations of wells.

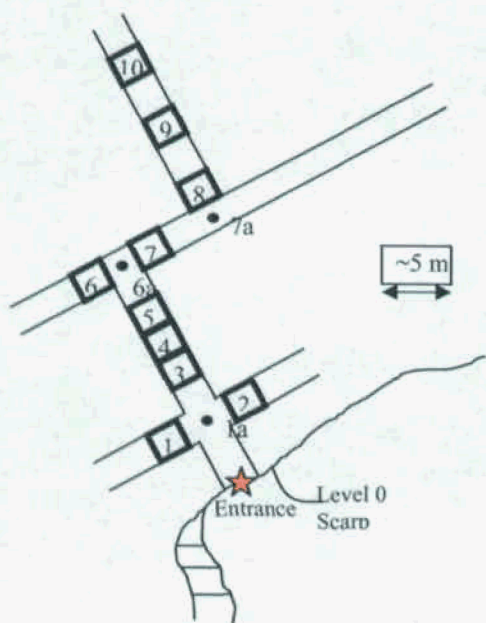


Fig. 2. Schematic map of unsaturated zone sample collection sites from the mine adit located on the +00 level.

progress. Radium was co-precipitated with BaSO_4 and short-lived daughter nuclides in equilibrium with Ra isotopes (^{226}Ra , ^{228}Ra , ^{224}Ra and ^{223}Ra) were counted by γ -spectrometry [1].

Uranium samples were divided into aliquots and spiked with ^{233}U tracer. After tracer equilibration, uranium was purified using anion exchange column chemistry and loaded onto single or triple Re filaments.

Uranium was measured multi-statically using a GV Sector 54 mass spectrometer equipped with multiple Faraday cups and a Daly ion counting detector. Daly/Faraday crossover was determined by switching the ^{235}U beam between these collectors, and instrumental mass fractionation was corrected by normalization to the natural $^{238}\text{U}/^{235}\text{U}$ ratio of 137.88. Measurement accuracy for both U concentration and $^{234}\text{U}/^{238}\text{U}$ ratio was verified by measurement of the NBS U-960 and NIST 4321C natural uranium standard reference materials. Filtration and process blanks were generally measured with each set of samples and are negligible in comparison to U sample size.

III. RESULTS

Uranium concentrations and $^{234}\text{U}/^{238}\text{U}$ ratios for saturated zone samples are given in Table 1. Decay counting measurements of Rn, Ra, Po and Pb isotopes are presented in Table 2.

IV. DISCUSSION

IV.A. Uranium constraints on groundwater flow in the saturated zone

In 2003, three groundwater wells were drilled directly adjacent to (PB1) and ~50 m on either side of the uranium deposit (PB2 and PB3) in order to evaluate the transport of U-series nuclides in three dimensions. After drilling, U concentrations were elevated in all of the three wells (0.1-18 mg U/kg) due to drilling activities. Some of the wells had pH values as high as 11.3 from detergents in the drilling fluids, and the interaction of these high pH fluids with the overlying uncased rock led to the high U

TABLE 1. Groundwater well uranium concentrations and $^{234}\text{U}/^{238}\text{U}$ activity ratios measured by mass spectrometry.

Well	Sample ID	Date Collected	[U] (ng/g)	$^{234}\text{U}/^{238}\text{U}$ (activity ratio)
PB-1	SPC1025028	1-Jul-03	802.3	1.029
	SPC1025022	6-Aug-03	328.0	1.058
	SPC1025029	29-Aug-03	166.5	1.021
	SPC1025044	9-Dec-03	241.4	1.011
	SPC1025047	9-Dec-03	194.9	1.028
	SPC1025058	5-Feb-04	92.96	1.006
	SPC1031344	17-Nov-04	43.88	1.044
	SPC1038245	11-Nov-05	32.82	1.010
	PB-2	SPC1025017	6-May-03	137.2
SPC1025026		1-Jul-03	87.28	1.019
SPC1025023		6-Aug-03	12.67	1.025
SPC1025030		29-Aug-03	16.94	1.019
SPC1025043		8-Dec-03	123.9	1.079
SPC1025059		5-Feb-04	79.16	1.039
SPC1031341		17-Nov-04	11.91	1.090
PB-3	SPC1025019	7-May-03	18200	1.794
	SPC1025027	1-Jul-03	3596	1.363
	SPC1025024	6-Aug-03	5343	1.516
	SPC1025046	10-Dec-03	966.5	1.835
	SPC1025060	5-Feb-04	820.6	1.974
	SPC1031350	17-Nov-04	540.6	1.969
	SPC1038246	11-Nov-05	369.9	1.966
PB-4	032703-1	27-Mar-03	0.162	1.422
	SPC1025020	7-May-03	0.482	1.712
	SPC1025025	6-Aug-03	2.388	1.494
	SPC1025031	29-Aug-03	1.110	1.631
	SPC1025048	12-Dec-03	0.091	1.470
	SPC1025061	5-Feb-04	15.41	1.048
	SPC1031127	8-Apr-05	3.197	2.012
	Pozos Ranch	030701-6	7-Mar-01	5.711
020403-1		4-Feb-03	5.024	2.156
SPC1025021		7-May-03	5.291	2.149
SPC1025039		26-Sep-03	5.097	2.150
SPC1031348		17-Nov-04	5.773	2.108
SPC1031118		6-Apr-05	5.135	2.159
SPC1038249		11-Nov-05	5.189	2.152
Pena Blanca Well	020403-3	4-Feb-03	10.02	4.603
	SPC1025037	26-Sep-03	9.945	4.603
	SPC1031114	6-Apr-05	9.538	4.616

Typical uncertainties (2σ) are <0.5% for [U] and <0.2% for $^{234}\text{U}/^{238}\text{U}$.
 Half-lives used are 4.468E9 yr for ^{238}U and 245250 yr for ^{234}U .

TABLE 2. Groundwater well short-lived uranium-series nuclide activities measured by decay counting.

Well ID	Rn-222 (dpm/L)	Ra-226 (dpm/m ³)	Ra-228 (dpm/m ³)	Ra-224 (dpm/m ³)	Ra-223 (dpm/m ³)	Pb-210 (dpm/m ³)	Po-210 (dpm/m ³)
Pena Blanca	1831 +/- 80	268 +/- 5	2614 +/- 69	4984 +/- 64	96 +/- 52	45.5 +/- 1.4	4.9 +/- 0.3
Pozos Ranch	1421 +/- 67	192 +/- 5	622 +/- 19	997 +/- 16	29 +/- 17	3.2 +/- 1.2	0.69 +/- 0.07
PB-4	595 +/- 37	716 +/- 7	540 +/- 14	589 +/- 10	29 +/- 20	94.1 +/- 4.3	95.3 +/- 3.7

Uncertainties are 1 σ based on counting statistics. Results are decay-corrected back to the sample collection dates of April 6 and 8, 2005 (see dates and sample numbers in Table 1).

concentrations. As shown in Figure 3, U concentrations in these wells have exponentially decreased over time.

Uranium isotopic mixing relationships are shown in Figure 4, in which conservative mixing between components results in linear trends. The ²³⁴U/²³⁸U activity ratios are similar for PB1 and PB2 (1.005 to 1.090) but higher for PB3 (1.36 to 1.97) over the 2003-2005 time period. These data, along with results of pumping and well conditioning [2], which found drawdown of PB1 during PB2 pumping and vice versa, suggest interconnectivity between the PB1 and PB2 wells but not PB3. Regional groundwater wells located up to several km from the deposit also have distinct U isotopic characteristics, indicative of multiple components of uranium. These components consist of 1) a high U component with ²³⁴U/²³⁸U activity ratios near unity, as would result from bulk dissolution of a rock in secular equilibrium, 2) components with high ²³⁴U/²³⁸U activity ratios and variable U concentration, as one would obtain for samples with a high recoil-related component and aqueous chemical fractionation of U, and 3) components with intermediate ²³⁴U/²³⁸U activity ratios and variable U concentration. The uranium chemical fractionation may be due to any number of processes including dissolution/precipitation and evaporation/dilution.

The decreasing uranium concentrations in the newly drilled wells can be modeled in a variety of ways. Here we present two models which place constraints on groundwater flow velocity. The first is a simple one-dimensional tank model, in which the tank consists of the saturated zone well volume that is continuously flushed with groundwater with an ambient, background U concentration. The second is a one-dimensional advection-dispersion model, in which groundwater movement causes U in the borehole to decrease, and dispersion causes a spread in tracer velocities and U retention in the borehole. In both of the models it is assumed that uranium is introduced as a slug to each of the wells, at t=0 and x=0, and transported as a conservative tracer in the direction of groundwater flow away from each of the wells. In the former model, the U concentration is a simple function of mean groundwater

flow velocity, whereas in the second model the relative U concentrations are dependent on the extent of dispersion as well as the mean groundwater flow velocity. If it is assumed that the background U in groundwater is negligibly small compared to the U defined by the initial slug, then the first model yields the following relationship derived for this study:

$$v = \frac{V}{2hr(t_2 - t_1)} \ln(c_1/c_2) \quad (1)$$

where v is the groundwater flow velocity, V is the saturated zone well volume, h is the casing perforation height, c_1 is the U concentration in the well at time t_1 and c_2 that at t_2 , and r is the well casing internal radius.

With a similar assumption for the advection-dispersion model, one obtains the following relationship for flow velocity [3]:

$$v = \sqrt{\frac{4Dh \ln((c_2/c_1)\sqrt{(t_2/t_1)})}{t_1 - t_2}} \quad (2)$$

with c_1 , c_2 , t_1 , t_2 the same as above and Dh the longitudinal hydrodynamic dispersion coefficient.

For both models, the most reliable velocity data will likely be obtained from the initial decrease in U concentration in 2003. In this situation, the models are relatively unaffected by the magnitude of the background or ambient U in groundwater or any significant U dissolution in the wells. Using the 2003 data for all three wells, the first model yields flow velocities which are in the range of 0.7-2.1 m/yr. For the advection-dispersion model, an independent relationship between velocity and dispersion is required, and these two parameters have been found to be correlated in both laboratory and field studies [4]. Using typical relationships between velocity and dispersion for laboratory and field studies along with the relationship between U concentration and time observed from our 2003 data, estimates of both velocity (~0.5-10 m/yr) and dispersion coefficient (0.00001 - 0.002 cm²/s) can be obtained from the results for all three wells (Figure 5).

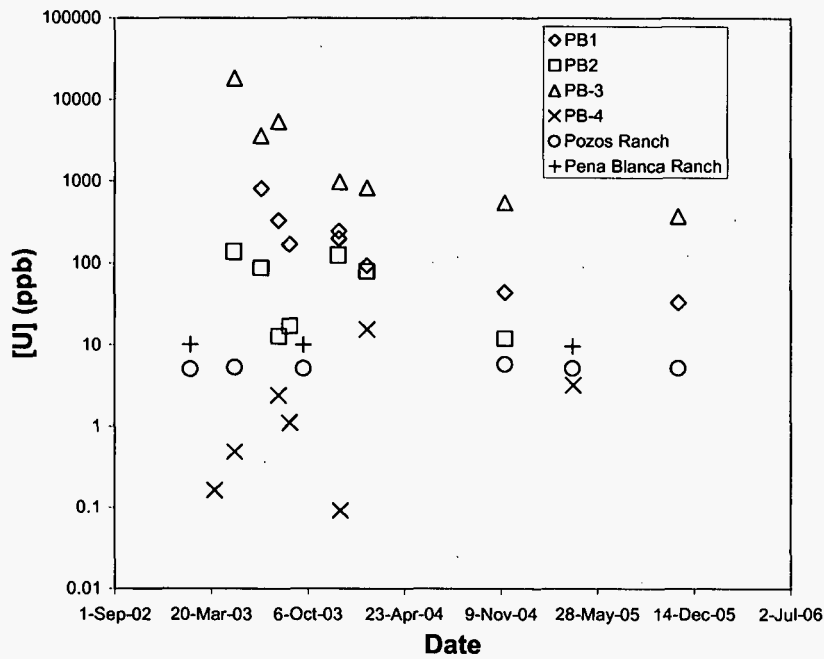


Fig. 3. Uranium time-series for wells. Uranium concentrations in newly drilled wells in 2003 (PB1, PB2, PB3) have generally exponentially decreased over time, although well pumping and conditioning in December 2003 disturbed the trend. Results for PB4, Pena Blanca Ranch, and Pozos Ranch have remained low over this time period.

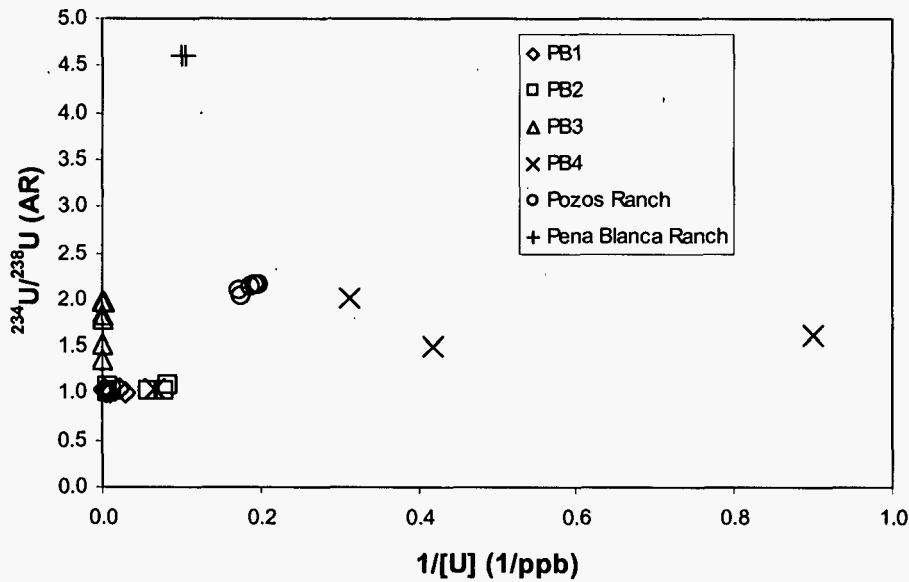


Fig. 4. Uranium isotope mixing diagram for the saturated zone. Multiple components for uranium, with limited mixing between these components, are indicated. An exception to this is the results for PB1 and PB2, which along with well pumping and conditioning results suggest interconnectivity between the groundwater in these two wells.

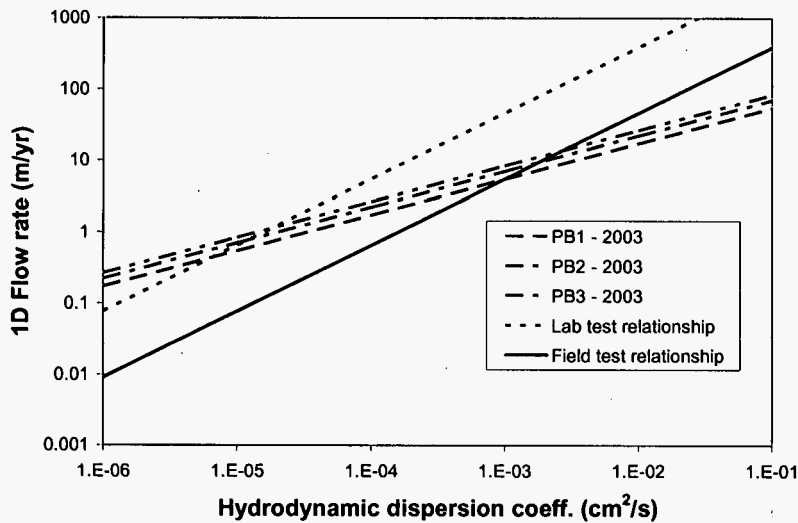


Fig. 5. 1-D advection/dispersion model results, indicating limited groundwater flow rates of ~ 0.5 to 10 m/yr. A tank model also suggests limited flow rates of 0.7 to 2.1 m/yr. The lab and field test relationships used to determine the dispersion-velocity relationship at this site are from [4].

The low flow velocities obtained from these data are consistent with interconnectivity between PB1 and PB2 observed during field tests [2], as much larger transient groundwater flow rates would be expected during active pumping of each of these wells. The flow velocities calculated above are those obtained under unperturbed, natural conditions. While our uranium concentration and isotopic results and the low productivity of these wells consistently suggest limited saturated zone groundwater flow and mixing, additional anticipated work with artificial tracers should better establish groundwater flow velocities and gradient at this site.

IV.B. Uranium transport in the unsaturated zone

The uranium isotopic systematics for unsaturated zone (UZ) groundwater at Peña Blanca can be evaluated using a model simulating a non-steady state situation for uranium isotope transport in groundwater. Applicable to unsaturated as well as saturated layers, the model provides constraints on in-situ radioisotope migration in dissolved and colloidal phases in terms of retardation factor and rock-water interaction (or water transit) time [1]. For uranium, the model is based on the fact that water passing through the UZ has its U concentration and $^{234}\text{U}/^{238}\text{U}$ ratio modified by U dissolution from rocks and α -recoil input of ^{234}U from rock surfaces. The model predicts that intermittent flushing of the UZ gives rise to waters having a linear relationship between reciprocal U concentration and $^{234}\text{U}/^{238}\text{U}$ ratio. For a linear array of

data, there are three possible interpretations of the slope and intercept. The most general of these interpretations is that the slope is a function of the non-flushing period and α -recoil rate of ^{234}U , whereas the intercept on the y-axis ($^{234}\text{U}/^{238}\text{U}$) varies with the relative rates of recoil to dissolution of uranium. If the recoil rate were to remain unchanged, the slope would increase as the non-flushing, dry-period lengthens, and a higher intercept would reflect a lower dissolution rate of U in the UZ. For situations where the intercept is near unity, the slope would also increase as the rock-water interaction time increases.

Uranium isotopic systematics for all of the adit waters collected from 1995-2005, including results obtained by Pickett and Murphy [5] are shown in Figure 6. From this plot, the uranium isotopic systematics appear to show a spatial dependence. Samples from the front of the adit generally have $^{234}\text{U}/^{238}\text{U}$ activity ratios near unity and higher U concentrations, as would result from dissolution of a rock in secular equilibrium. Samples from the rear of the adit are characterized by high $^{234}\text{U}/^{238}\text{U}$ activity ratios ranging from 2 to 5 and variable U concentration, as one would obtain for samples with a high recoil-related component and U concentration affected by varying U dissolution rates and/or rock-water interaction times. Finally, samples from the middle of the adit appear to be a mixture of these two endmembers. Based on all of the data obtained thus far, the uranium isotopic systematics in the UZ do not appear to show significant seasonal dependence.

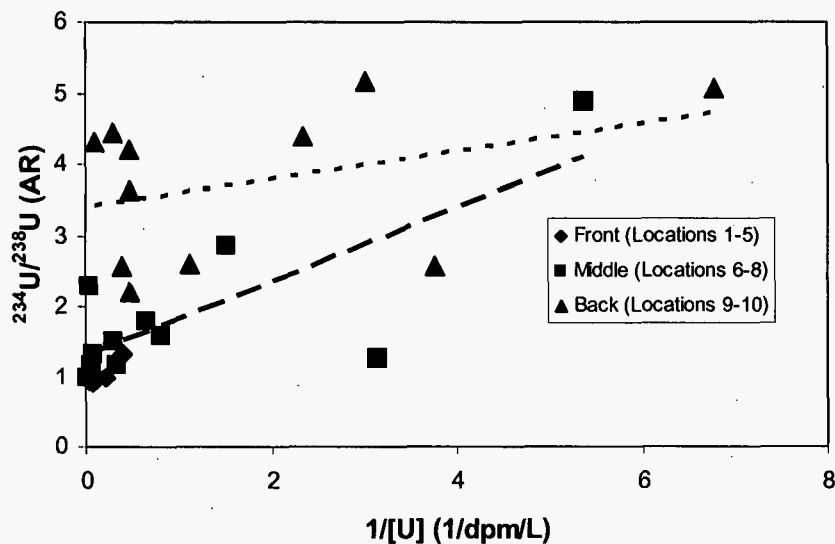


Fig. 6. Uranium isotope mixing diagram for the adit water samples from the unsaturated zone. A spatial dependence for the uranium isotopic systematics is indicated. Long and short dashed lines are the linear regressions for samples from the middle and back of the adit, respectively. Low intercept and high slope for samples from the front and middle of the adit are indicative of high uranium dissolution rates and long rock-water interaction times. Samples from the back of the adit have experienced lower uranium dissolution rates and variable but generally shorter rock-water interaction times.

The isotope mixing relationships seen in Fig. 6 appear scattered, and a linear model can only approximate the results for each location in the adit. However, the intercept and slope of the linear approximation reveals some consistent results in terms of uranium dissolution rates and/or rock-water interaction times. Samples from the front and middle of the adit correspond to low intercept (near secular equilibrium), reflecting increased U dissolution rates toward the front of the adit, where the ore deposit is located. Their generally high U concentration and slope reflect longer rock-water interaction times. The high intercept, low slope, and generally lower U concentration for samples from the back of the adit suggest that these samples have experienced decreased U dissolution rates and variable but generally shorter rock-water interaction times. Consistent with these results, a shorter seepage time has been observed at the back of the adit relative to the front of the adit (P. Dobson, pers. comm.).

Some of the scatter in the data for a given collection period could be due to the adit water collection system. Several factors could have contributed to the data scatter. Among the important ones are that we did not sample

waters representing discrete major rain events, and that the collection system allowed evaporation to have taken place. An improved collection system has been installed, which will hopefully remove any sampling artifacts and facilitate our future model validation effort.

IV.C. Short-lived uranium-series radionuclide transport in the saturated zone

Preliminary results (Table 2) show that the activities of the radioisotopes ^{228}Ra , ^{226}Ra , ^{224}Ra , ^{223}Ra , ^{210}Pb , and ^{210}Po are several orders of magnitude lower than that for ^{222}Rn , leading to calculation of high retardation factors for these three elements. Based on the uranium-series transport model of [1], we estimate that the in-situ retardation factors are $\sim 10^3$ for Ra and $\sim 10^4$ to 10^7 for Pb and Po. Among the three wells studied, retardation factors for Ra, Pb and Po are highest for the Pozos Ranch sample. Pozos Ranch water also appears to have the lowest concentration of colloids of the wells sampled in the area (A. Abdel-Fattah, pers. comm.). The high retardation factors of Pb and Po at Pozos Ranch may be related to the low abundance of colloids, if colloid-facilitated radionuclide transport in the groundwater is

important for these elements. Further measurements are currently being completed in order to evaluate the importance of colloid-facilitated transport for particle-reactive elements such as thorium.

There are two important sources of radon in groundwater: (1) direct input via α -recoil from aquifer solids and (2) in-situ production from decay of ^{226}Ra in the sorbed phases on the fracture surface of the solids. Our model suggests that for wells such as PB4, where the ^{222}Rn activity is low (Table 2), the input of ^{222}Rn to groundwater via α -recoil is an unimportant source compared to that from decay of the sorbed ^{226}Ra on the fractures. This suggestion is consistent with our measurements of the radium isotopic activities. As seen from Table 2, $^{224}\text{Ra}/^{228}\text{Ra}$ is close to unity in well PB4 and significantly greater than unity in wells Pozos Ranch and Peña Blanca. Similarly, $^{224}\text{Ra}/^{223}\text{Ra}$ is close to their equilibrium ratio of ~ 20 in well PB-4 but significantly greater than this equilibrium ratio in wells Pozos Ranch and Peña Blanca. The lack of excess ^{224}Ra in well PB4 clearly suggests that radionuclide input via α -recoil at this site is not important, likely due to the occurrence of large fractures in the limestone aquifer rocks. Applying the measured ^{222}Rn activities to our model, we estimate that the mean fracture widths in the aquifer rocks range from 0.2 to 4 μm in the three wells, with the largest fracture width ($\sim 4 \mu\text{m}$) occurring at well PB4.

Results for the longer-lived ^{226}Ra in the three wells can be compared with prior measurements by mass spectrometry reported by Goldstein et al. [6] for Pozos Ranch. The value for Pozos Ranch reported in Table 2 corresponds to a ^{226}Ra concentration of 0.087 fg/g, which is similar to prior mass spectrometric measurements of 0.086 and 0.124 fg/g for samples collected in 2000 and 2001. $^{226}\text{Ra}/^{238}\text{U}$ activity ratios for all three wells are low (0.04-0.30), in agreement with prior measurements for Pozos Ranch. Prior measurements of high $^{226}\text{Ra}/^{238}\text{U}$ activity ratios of 4 to 113 [6] and $^{226}\text{Ra}/^{230}\text{Th}$ activity ratios of 24 to ~ 10000 in adit water samples indicated preferential mobility of radium relative to uranium and thorium in UZ water near the deposit. This is most likely associated with radium mobility in fracture-filling materials near the deposit observed by Murrell et al. [7].

V. CONCLUSIONS

Uranium-series data for groundwater samples from the vicinity of the NOPAL I uranium ore deposit are being used to place constraints on rock-water interaction times in the unsaturated zone as well as groundwater flow rates and relative rates of mobility for various radionuclides in the saturated zone. Such data place useful constraints on processes and models of radionuclide transport, both at this site and by analogy at

Yucca Mountain. The radionuclide transport results will be incorporated in a Peña Blanca total system performance assessment model, and this will provide useful testing and validation for a similar performance assessment model at Yucca Mountain. In addition to these issues related to the Yucca Mountain repository, these results provide enhanced understanding of the rates of groundwater hydrologic processes in arid environments.

VI. ACKNOWLEDGMENTS

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