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THE PEÑA BLANCA NATURAL ANALOGUE PERFORMANCE ASSESSMENT MODEL

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Abstract – The Nopal I uranium mine in the Sierra Peña Blanca, Chihuahua, Mexico serves as a natural analogue to the Yucca Mountain repository. The Peña Blanca Natural Analogue Performance Assessment Model simulates the mobilization and transport of radionuclides that are released from the mine and transported to the saturated zone. The Peña Blanca Natural Analogue Model uses probabilistic simulations of hydrogeologic processes that are analogous to the processes that occur at the Yucca Mountain site.

The Nopal I uranium deposit lies in fractured, welded, and altered rhyolitic ash flow tuffs that overlie carbonate rocks, a setting analogous to the geologic formations at the Yucca Mountain site. The Nopal I mine site has the following characteristics as compared to the Yucca Mountain repository site.

- Analogous source: UO_2 uranium ore deposit = spent nuclear fuel in the repository
- Analogous geologic setting: fractured, welded, and altered rhyolitic ash flow tuffs overlying carbonate rocks
- Analogous climate: Semiarid to arid
- Analogous geochemistry: Oxidizing conditions
- Analogous hydrogeology: The ore deposit lies in the unsaturated zone above the water table.

The Nopal I deposit is approximately 8 ± 0.5 million years old and has been exposed to oxidizing conditions during the last 3.2 to 3.4 million years. The Peña Blanca Natural Analogue Model considers that the uranium oxide and uranium silicates in the ore deposit were originally analogous to uranium-oxide spent nuclear fuel. The Peña Blanca site has been characterized using field and laboratory investigations of its fault and fracture distribution, mineralogy, fracture fillings, seepage into the mine adits, regional hydrology, and mineralization that shows the extent of radionuclide migration.

Three boreholes were drilled at the Nopal I mine site in 2003 and these boreholes have provided samples for lithologic characterization, water-level measurements, and water samples for laboratory analysis of the saturated zone water chemistry. The results of the field investigations and laboratory analyses of rock and water samples collected at Nopal I are used to calibrate the Peña Blanca Natural Analogue Model.

I. INTRODUCTION

The Nopal I uranium deposit, located in the Sierra Peña Blanca, approximately 50 km north of Chihuahua City, Chihuahua, Mexico, is a natural analogue to the Yucca Mountain repository [1], and is referred to as the Peña Blanca natural analogue. Murphy and Codell [2] proposed that the uranium oxide and uranium silicates comprising the bulk of the radionuclides at the deposit were originally directly analogous to spent nuclear fuel, which is largely composed of uranium oxide. The Peña Blanca Natural Analogue Model (PBNAM) adapts the Yucca Mountain performance-assessment model to simulate the release and transport of radionuclides from the Nopal I ore deposit using GoldSim software [3]. The PBNAM is supported by field investigations and the results of laboratory analysis of rock and water samples collected at the Peña Blanca Nopal I ore deposit.

I.A Setting

The Nopal I ore deposit lies on the southeast side the Sierra Peña Blanca in a high desert terrain in the Basin and Range geologic province (Figures 1 and 2) [4]. The deposit lies in fractured, welded, and altered rhyolitic ash-flow tuffs similar to the volcanic rocks at Yucca Mountain [5]. The land surface at the Nopal I deposit exposes a small portion of the ore body.

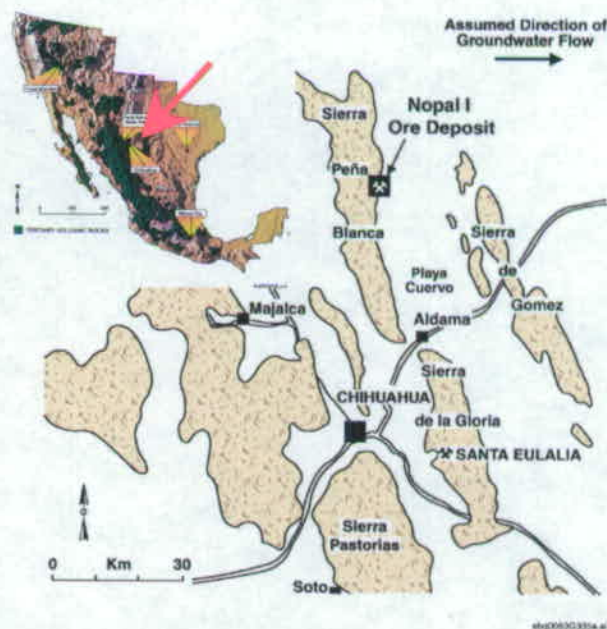


Figure 1. Location of the Nopal I Ore Deposit

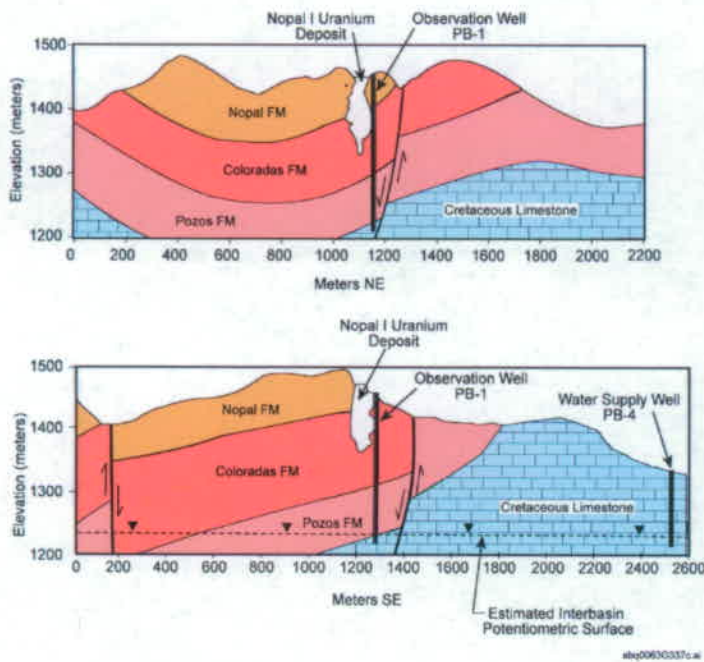


Figure 2. Cross sections of the Nopal I ore deposit and local geology

Mining of the Nopal I ore deposit in the late 1970s and early 1980s, left two prominent benches across the ore body at the 0-m and +10-m levels (vertical mine coordinates). There are two adits from land surface, and one principal shaft and adits at the 20-, 40-, and 70-m levels below the 0-m adit, as shown on Figure 3 (Figure 25 in [6]). The total depth of the main mineshaft is approximately 110 m below the -20-m level.

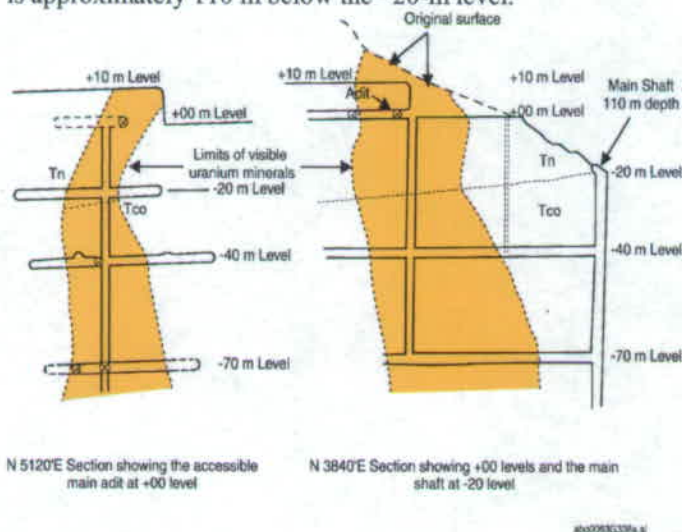


Figure 3. Nopal I Mine Shaft Schematic (Modified from [6], Figure 25, p. 205)

The Nopal I ore deposit was estimated to be approximately 8 ± 0.5 million years old ([7], p. 117). Examination of the weathering mineralogy of the ore body indicates that the deposit was stable and under reducing conditions until approximately three million years ago. At that time it was exposed to oxidizing groundwater, infiltration from precipitation, and weathering processes resulting in an alteration zone of oxidized secondary minerals, including uranium silicates, around the ore body [2].

The top 30 m of the ore deposit lies in the Nopal Formation, a fractured rhyolitic tuff, and the lower 70 m lies in the Coloradas Formation, a weakly welded, fractured, ignimbritic tuff. The ore deposit is underlain by 60 m of Pozos Formation, a silicified, detrital conglomerate, composed principally of altered limestone clasts, which lies on Cretaceous limestone. A near vertical fault, with greater than 10 m of offset, intersects the hill containing the ore body and lies to the east of the deposit, but does not appear to cut the ore body. Minor faults and fractures are observed in the vertical walls of the open faces of the mine above the 0- and 10-m levels.

The ore body has a roughly cylindrical, breccia-pipe-like form, approximately 18 m by 30 m in the horizontal plane and 100 m in the vertical dimension. The deposit is presently estimated to contain 333 mT of uranium [5]. Uranium comprises approximately 0.23 percent of the deposit by volume [5]. Percy et al. [4] and Murphy and Codell [2] estimated that the ore body contained 408 mT of uranium as uranium oxide before it was oxidized.

The Nopal I ore deposit lies in the unsaturated zone. The water table is approximately 122 m below the ore deposit ([9], Section 10.4)). Green and Rice [10] conducted an artificial recharge study at the Nopal I mine, and the field observations were used to estimate the hydraulic properties of the intact and oxidized portions of the ore body. Their investigations indicate that the total porosity of the ore deposit ranges from 0.05 to 0.08 for unaltered rock, to 0.30 for altered rock. The saturated hydraulic conductivity ranges from 6×10^{-12} cm/s for the unaltered rock, to 1×10^{-7} cm/s for the altered rock.

I.B Conceptual Model

The PBNAM considers the possibility of the generation, liberation, and transport of ^{99}Tc and uranium species from the Nopal I deposit, as well as protactinium, radium, and thorium. The PBNAM considers a steady-state condition in which the magnitude of the source term with respect to time depends on the degradation rate. The

unsaturated and saturated zones are divided into mixing cells to simulate radionuclide transport.

The conceptual model for radionuclide transport from the Nopal I ore body assumes that ^{99}Tc has been generated primarily by the spontaneous fission of ^{238}U [11]. The ^{99}Tc inventory for the simulations was developed based on the potential production of ^{99}Tc from the spontaneous fission of ^{238}U . The estimation is based on the algorithm presented in [11] and the abundance of uranium species at the Nopal I deposit.

The ^{99}Tc produced from ^{238}U is conservatively [12] presumed to be in the soluble and mobile form of pertechnetate ion (TcO_4^-) [13]. As such it is subject to leaching from the Nopal I ore body and available for transport by groundwater. The work of [14] indicates that uranium-mineral alteration of the Nopal I ore deposit has only resulted in local-scale migration of uraniferous species with precipitation of secondary uranium minerals and sorption of released uranyl species after tens of meters of lateral travel. Given the age of the Nopal I deposit, the PBNAM analysis uses ^{99}Tc as a surrogate radionuclide to estimate transport of conservative, non-sorbing species in the natural environment.

The PBNAM of the Nopal I ore deposit treats the original and present uraninite in the ore body as having been subject to oxidation and dissolution following the change from reducing to oxidizing conditions approximately three million years ago. Dissolution was followed by the release of uranium species in a manner similar to the degradation of the spent-fuel waste forms in the waste packages expected to be emplaced in the Yucca Mountain repository. However, the PBNAM has no waste package to delay water-uraninite interaction or to hinder direct release and transport of radionuclides. In the PBNAM, the dissolved species are subject to advective and diffusive transport from the ore body.

II. MODEL CONFIGURATION

The PBNAM was configured based on the physical description of the Nopal I mine site found in [4] and [15]. The physical description of the site was supplemented with field observations and additional information provided during site visits. Field observations obtained during drilling and sampling operations at observation wells PB-1, PB-2, and PB-3 (Figure 4), conducted in 2004 and 2005, were also used.

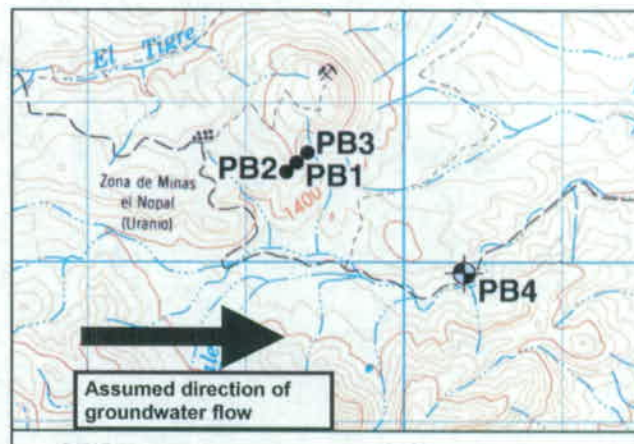


FIGURE 4. LOCATION OF OBSERVATION WELLS

The PBNAM is analogous to the overall structure of the Yucca Mountain total-system performance assessment model. The analogous performance measure for the PBNAM is the agreement of radionuclide concentrations predicted in groundwater in the saturated zone beneath and downgradient from the Nopal I ore body with observed concentrations in groundwater samples obtained from wells at and near the Nopal I mine.

II.A Initial/Boundary Conditions

A one-dimensional flow tube type model was used for the PBNAM. The upstream model boundary is defined by precipitation infiltration. The climate of the Peña Blanca region is arid, with an estimated 250 mm/yr of precipitation [4]. Perched water in shot holes drilled into the +10-m level indicates that precipitation infiltration occurs in the area of the Nopal I mine [16]. Because of the climatic similarity of Sierra Peña Blanca and the Yucca Mountain site, and because there are no detailed infiltration studies at the Nopal I mine site, the precipitation and infiltration observations made at Yucca Mountain are used to constrain the PBNAM [17]. The PBNAM assumed a constant average infiltration rate of 6 mm/yr. Transient climate fluctuations were not considered and would not likely influence the long term model performance. The downstream model boundary was taken to be an infinite volumetric sink.

Initial water saturation was set to zero and one in the UZ and SZ respectively. These initial saturation values do not affect the steady-state model performance examined here. Initial radionuclide concentrations are defined for the ore body.

II.B Source Term Dissolution and Radionuclide Inventory

The source term for the PBNAM was an ovoid cylinder of uranium oxide based on the dimensions estimated from the mine workings (Figure 3). The source was defined using a single source term group in the PBNAM consisting of two waste packages. This methodology was used to allow the definition of a dissolution rate for the uranium oxide ore from time zero, which is defined as the time when the ore body was exposed oxidizing conditions approximately three million years ago.

Uranium-oxide degradation was modeled using the same rate equation used in the Yucca Mountain performance-assessment model. The dependent variables of the dissolution rate of the ore body were assumed to be pH, total carbonate concentration, temperature, and oxygen fugacity. The commercial spent fuel (CSNF) dissolution rate model developed in [18] is

$$\text{Log (DR)} = a_0 + a_1 \times T^{-1} + a_2 \times \text{pCO}_3 + a_3 \times \text{pO}_2 + a_4 \times \text{pH} \quad (1)$$

where

- DR is the dissolution rate
- a_0 , a_1 , a_2 , a_3 , and a_4 are uncertain regression parameters in the PBNAM
- T is temperature
- pCO_3 is the negative log of total carbonate molar concentration (uncertain parameter)
- pO_2 is the negative log of oxygen fugacity
- pH is the negative log of hydrogen ion concentration (uncertain parameter).

The sequence of uraninite alteration at Nopal I is modeled as similar to that of CSNF and UO_2 in the laboratory tests. The corrosion products observed in laboratory CSNF and UO_2 tests conform to the mineral phases seen at Nopal I [18].

The inventory of uranium species and some daughter products, together with radiochemical analyses of Nopal I ore piles and vegetation residues [19], were used to estimate the approximate inventory of radionuclides used for the PBNAM simulations.

The estimated radionuclide inventory was used as the initial inventory for the PBNAM. Because of its half-life, ^{99}Tc was further defined with an accumulation function [11] based on a steady-state inventory of ^{238}U , which has a half-life of 4.5×10^9 years. ^{99}Tc is assumed to grow throughout the model domain. Using the assumed

inventory, the dissolved mass from the ore deposit was mixed into a single mixing cell, with the volume set equal to the physical volume of the ovoid cylinder representing the uranium deposit, taking into account the porosity [10]. The total flow out of the mixing cell was set equal to the infiltration flux times the cross-sectional area of the deposit. The PBNAM incorporates solubility values for ^{234}U , ^{235}U , ^{238}U , ^{231}Pa , ^{232}Th , ^{226}Ra , and ^{99}Tc that fall within the range of values used in the Yucca Mountain performance-assessment model.

II.C Unsaturated Zone

The vertical UZ section of rock below the ore body is approximately 120 m thick ([6], Table 12, Section V, p. 213; and [9], Section 10.4) and is composed both of the lower part of the volcanic tuff and volcanic conglomerate of the Pozos Formation and of the upper part of the Cretaceous limestone that underlies this region of northern Mexico. Because there were not sufficient field observations from the Nopal I site to create a sophisticated site-specific model, transport of ^{99}Tc through the UZ beneath the Nopal I deposit was modeled using mixing cells in the GoldSim software. In the Peña Blanca Natural Analogue Model, infiltrating water contacted the ore body and the volume of percolating water corresponded to that derived from the present-day climate. The 120-m thick UZ was discretized into five mixing cells. The mass release from the ore deposit in the source-mixing cell was fed into the first UZ mixing cell at a rate equal to the infiltration rate times the cross-sectional area. The cross-sectional area of the ore body (18 m by 30 m) was used, with the height of the deposit (100 m) [4] and a porosity of 15 percent [10] used to estimate the volume of fluid percolating through the deposit.

II.D Saturated Zone

The PBNAM accumulates the water flowing through the unsaturated zone and passes it to the saturated zone at the water table. The modeled fluid contains radionuclides leached from the Nopal I ore body based on the estimated radionuclide inventory for the Nopal I ore body. The estimated leachate from the Nopal I deposit primarily contains uranium isotopes and ^{99}Tc . The theoretically contaminated groundwater flows eastward in the saturated zone through the Cretaceous limestone according to the hydraulic gradient ([20], Figure 4.8). Saturated zone flow in the PBNAM uses a 1-D flow and transport model with cross-sectional area assumed to be identical to the ore body. The saturated zone was discretized into ten mixing cells arranged in series consistent with a 1-D model. The rate of mass flux in the saturated zone was set equal to the mass flux leaving the ore body. The PBNAM captures

the concentrations provided by the saturated zone mixing cell at any specified distance of interest, such as a monitoring well. Observation wells PB-2 and PB-3 are 50 m from PB-1, the approximate center of the Nopal I ore deposit. The PBNAM did not consider dose-to-receptor values because the goal of the investigation was to estimate concentrations in groundwater of ^{99}Tc and some uranium species at selected distances from the Nopal I mine.

III Results

Figure 5 shows the results of the base-case PBNAM simulation. The concentration of each of the radionuclide species simulated is shown for the model cell directly beneath the ore body. The results were calibrated using concentrations of uranium reported for water samples collected from boreholes PB-1, PB-2, and PB-3 installed at the Nopal I ore deposit in 2003. Figure 5 shows the results of the base-case simulations using the best estimates of site parameters and capture of the radionuclide concentrations at the unsaturated-saturated zone interface beneath the ore deposit. The results show that the radionuclide concentrations are dominated by ^{238}U , with lesser contributions from other uranium species. The concentration of ^{231}Pa peaks and falls off because of the limited protactinium inventory. With its half-life of 3.25×10^4 years, the PBNAM does not include a generation function for ^{231}Pa . The results also show that the 2.13×10^5 year half-life of ^{99}Tc means that ^{99}Tc rapidly decays, producing only very low concentrations for the majority of the simulation time.

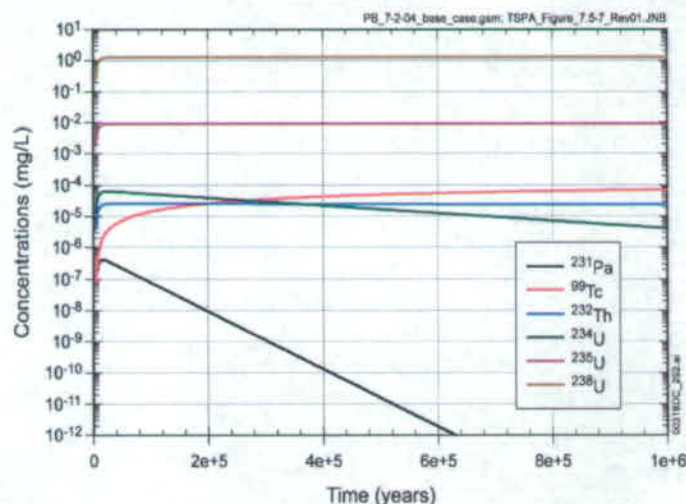


Figure 5. Calculated Radionuclide Concentrations at the Top of the Saturated Zone beneath the Nopal I Ore Deposit

After the initial increase in concentration, the concentration of ^{99}Tc is maintained by taking into account ^{99}Tc production from spontaneous fission of ^{238}U [11]. Because of the 4.5 billion year half-life of ^{238}U , the resulting ^{99}Tc concentrations did not noticeably increase or decrease for the remainder of the simulation period but remained at a relatively low concentration throughout the entire one million-year simulation period.

The results were investigated using sensitivity analysis for the distribution coefficient (K_d). Figure 6 shows the sensitivity of the results to the K_d of material in the unsaturated zone. In this simulation, a relatively large K_d value was used for uranium ($1 \text{ m}^3/\text{kg}$), but the K_d for ^{99}Tc was set at zero. The results indicate that the ^{238}U concentrations were reduced, whereas the ^{99}Tc concentrations remained at a constant low concentration.

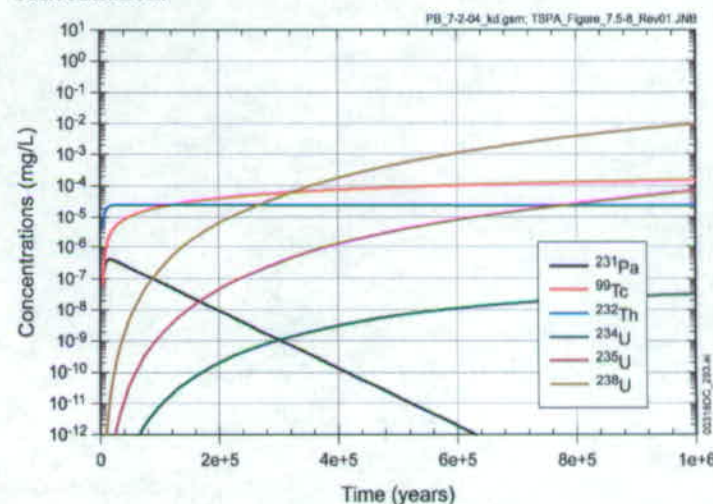


Figure 6. Calculated Radionuclide Concentrations beneath the Nopal I Ore Deposit Using High K_d for ^{238}U

Figure 7 shows the observed concentrations of uranium reported for water samples collected from observation wells PB-1, PB-2, and PB-3 (Figure 4), and from the mining camp supply well, PB-4 (Figure 4), as reported in Section 10.4 of [9]. The high initial concentrations of uranium in PB-1, PB-2, and PB-3 are likely due to drilling contamination. The gradual decay of concentrations seen in these boreholes reflects post-drilling dilution by groundwater flow through the boreholes. Therefore, the late-time sample analyses on Figure 7 are more representative of natural concentrations. These late-time sample analyses were used to calibrate the results obtained with the PBNAM. As seen on Figure 7, uranium concentrations at the mining-camp well are negligible.

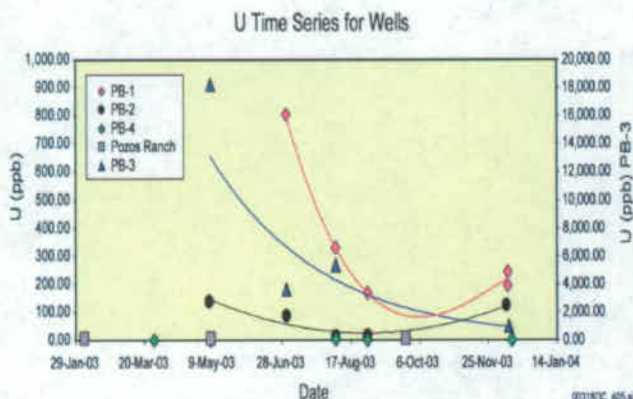


Figure 7. Uranium Concentrations in Groundwater in Water Samples from Nopal I Observation Wells. PB-1, PB-2, and PB-3 Drilled in Fiscal Year 2003 ([9], Figure 10.4-15)

Figure 8 shows the results of the base-case simulation for ^{238}U for 100 realizations of the uncertain dissolution parameters, but not including sorption. The observed concentrations of the late time sample analyses from observation wells PB-1, PB-2, and PB-3 (Figure 4) are also shown on Figure 8. The observed concentrations beneath the ore deposit are bracketed by the range of results obtained in the simulations and within the uncertainty of the source dissolution parameters

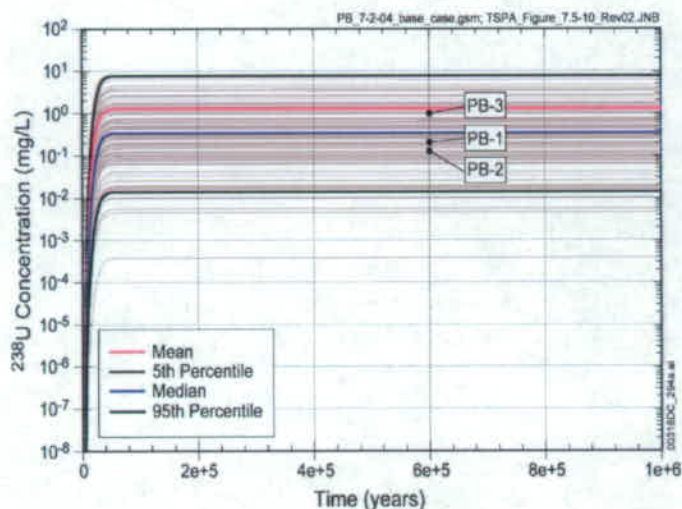


Figure 8. Results of the Base-Case Simulation for ^{238}U for 100 Realizations of the Uncertain Dissolution Parameters with Observed Concentrations in Late Time Groundwater Samples from the PB-1, PB-2, and PB-3 Boreholes

IV. CONCLUSIONS

The Nopal I uranium-ore deposit at the Sierra Peña Blanca was modeled as a natural analogue of the Yucca Mountain repository to estimate the migration of uranium species and analyzable quantities of dissolved ^{99}Tc using the PBNAM.

The analysis indicates that picogram quantities of ^{99}Tc generated by spontaneous fission of ^{238}U may be detectable at 50 m and 1,300 m from the Nopal I ore deposit. In addition, uranium appears to be transported in limited quantities. However, released uranium is apparently exchanging with uranium minerals that precipitate in fractures around the ore deposit. Despite this precipitation, there is sufficient uranium available to be transported through the Cretaceous limestone below the Sierra Peña Blanca.

The results at the distances of the Nopal I boreholes show sorption of uranium because of the values of the uranium distribution coefficient used in the PBNAM. The results indicate that uranium sorption may increase the estimated concentration of ^{99}Tc relative to the uranium concentration. However, the results indicate that the ^{99}Tc concentration would still be very low, but analyzable at a concentration of 2.8×10^{-5} mg/L. The low concentrations of uranium at the Nopal I boreholes may be due either to uranium retardation in the vicinity of the ore deposit or to the location of the boreholes relative to the regional flow direction.

Radionuclide transport by groundwater is the most likely off-site transport pathway that could possibly affect the performance of the Yucca Mountain repository. Despite some uncertainty in the estimated direction and gradient of groundwater flow, the Peña Blanca natural analogue site offers a unique opportunity to examine the groundwater flow and transport of uranium and some of its daughter products in a climatic and geologic setting very similar to that of Yucca Mountain. Both sites are set in volcanic tuff in an oxidizing unsaturated zone, and they are in similar semiarid environments. The Nopal I mine was originally comprised of uraninite, which is essentially the same material as the spent nuclear fuel destined for the Yucca Mountain repository, and the ore deposit was analyzed using a modified version of the metal-fuel dissolution model used in the analysis of the performance of the repository. The PBNAM evaluated the transport of ^{99}Tc , an expected conservative ion that will be released from the waste packages in the Yucca Mountain repository. The calculated values of uranium concentration in the groundwater beneath the Nopal I ore body are generally in agreement with observed concentrations (Figure 7). Thus, the PBNAM simulation

demonstrates the ability of the Yucca Mountain performance-assessment model to forecast the mobilization and transport of radionuclides from a geologic source analogous to the nuclear material emplaced at the Yucca Mountain repository.

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