GAMMA-RAY CHARACTERIZATION OF THE U-SERIES INTERMEDIATE DAUGHTERS FROM SOIL SAMPLES AT THE PEÑA BLANCA NATURAL ANALOG, CHIHUAHUA, MEXICO.

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by

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ABSTRACT

The Nopal I uranium deposit is located in the Sierra Peña Blanca, Mexico. The deposit was mined in the early 1980s, and ore was stockpiled close by. This stockpile area was cleared and is now referred to as the Prior High Grade Stockpile (PHGS). Some of the high-grade boulders from the site rolled downhill during stockpiling; for this study soil samples were collected from the alluvium surrounding and underlying one of these boulders. A bulk sample of the boulder was also collected. Because the Prior High Grade Stockpile had no ore prior to the 1980s, a maximum residence time for the boulder is about 25 years; this means that the soil was at background as well. The purpose of this study is to characterize the transport of uranium series radionuclides from ore to the soil.

Transport is characterized by determining the relative activities of individual radionuclides and daughter to parent ratios. Isotopes of the uranium series decay chain detected include ^{210}Pb, ^{234}U, ^{230}Th, ^{226}Ra, ^{214}Pb, and ^{214}Bi. Peak areas for each isotope are determined using gamma-ray spectroscopy with a Canberra Ge (Li) detector and GENIE 2000 software.

The boulder sample is close to secular equilibrium when compared to the standard BL-5 (Beaver Lodge Uraninite from Canada). Results for the soils, however, indicate that some daughter/parent pairs are in secular disequilibrium. These daughter/parent (D/P) ratios include $^{230}Th/^{234}U$, $^{226}Ra/^{230}Th$, and $^{210}Pb/^{214}Bi$. The gamma-ray spectrum for organic material lacks ^{230}Th peaks, but contains ^{234}U and ^{226}Ra. The results, combined with previous studies require multistage history of mobilization of the uranium series radionuclides. Earlier studies at the ore zone could only limit the time span for mobilization to a few thousand years. The contribution of this study is that the short residence time of the ore at the Prior High Grade Stockpile requires a time span for mobilization of 20-30 years.

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1. INTRODUCTION

The Peña Blanca natural analog is located in the Sierra Peña Blanca, approximately 50 miles north of Chihuahua City, Mexico. The Peña Blanca site is considered a natural analog to the proposed Yucca Mountain nuclear waste repository because they share similar characteristics of structure, volcanic lithology, tectonic activity, and hydrologic regime.

One of the uranium-mineralized zones at Peña Blanca is the Nopal I mine. This deposit lies mostly in the Tertiary Nopal formation, which is silicic ash flow tuff. The uranium bearing deposit is part of a large breccia pipe in ignimbrites dated to be 44 Ma (Alba and Chavez 1974). Previous studies focused on the brecciated zone and the surrounding area (Pearcy et al. 1994, Pearcy et al. 1995, Prikryl et al. 1997, Leslie et al. 1999, Murrell et al. 1999, and Wong et al. 1999). Most samples that were analyzed for these studies were infillings from several fractures that make up an E-W fracture zone that runs from the middle of the breccia zone to the non-brecciated country rock (Fig.1).

The focus of this study is the mobility of radionuclides from high-grade ore boulders of the Prior High Grade Stockpile into local soils. The Prior High Grade Stockpile was located approximately 250 meters west of the Nopal I mine. High-grade ore from the mine was stockpiled here in the 1981 and boulders rolled down hill onto uncontaminated ground. The stockpile was later moved to another location in the 1993. The advantage of this is a short, well constrained residence period. One of the boulders that rolled down hill is the focus of this study. This situation presents a unique opportunity to analyze the mobility of the radionuclides from this boulder into the soil, to accomplish this soil samples were collected under, around, and into the subsurface near the boulder.

The soil samples were then analyzed using gamma ray spectroscopy. This method was used because individual radionuclide peaks from the uranium decay series could be produced and relative activities determined. Then, these peaks could be used to form ratios using daughter/parent pairs to determine equilibrium or disequilibrium for that pair. By knowing the characteristics of each radionuclide and this ratio, a deficiency or excess could be determined for either the daughter or parent.

Substantial portions of this study are published in a CD volume from the International High Level Radioactive Waste Management Conference in Las Vegas, NV in May 2006 (French et al. 2006). The publication was prepared and edited by French.

2. GEOLOGIC SETTING

The Sierra Peña Blanca is located about 50 miles north of Chihuahua City, Chihuahua, Mexico. The Sierra Peña Blanca is a horst block in a basin and range province known as the Chihuahua tectonic belt that contains many northwest striking faults (Goodell 1981). Before this region experienced extension, it was part of an inland sea that covered parts of Mexico and the southwestern United States. Therefore, many carbonate sequences can be found at the Sierra Peña Blanca. Following the retreat of the inland sea, Laramide compression initiated volcanism creating the volcanic stratigraphy of tuff sequences that cover a majority of Sierra Peña Blanca (Reyes-Cortez 1997).

Uranium at the Nopal I mine is located in a brecciated zone about 40 meters in diameter and at least 100 meter thick that occurs at the intersection of two small step faults (Altamirano 1992; Pearcy et al. 1994; Reyes-Cortes 1997; Goodell 1981). This was thought to be a magmatic breccia pipe or subsurface conduit that lies in the Nopal and Coloradas Formation tuffs. These two formations lie unconformably above a limestone conglomerate called the Pozos Formation. Whatever the ultimate origin of the brecciated zone, the breccia allowed hydrothermal waters to travel through conduits and precipitate uranium in fractures or voids (Wong 1994). In the early history of the deposit, reducing conditions caused U^{+4} to be precipitated producing uraninite, but more recently water and other hydrothermal processes have caused the uraninite to oxidize forming minerals like weeksite and uranophane.

There are many similarities that make Peña Blanca an excellent natural analogue to Yucca Mountain. Yucca Mountain is a horst block in a region of basin and range dominated by strike-slip and intrablock faulting (Day et al. 1998). Yucca Mountain is also composed of sequences of welded tuffs and non-welded tuffs from calderas that lie to the north, which are

rhyolitic in composition (Day et al. 1998). The unit selected to hold the proposed high-level nuclear waste repository is the Topopah Spring member of the Paintbrush Tuff, which is a welded tuff in the unsaturated zone (www.ocrwm.doe.gov/documents).

Both sites are semi-arid much like the rest of northern Mexico and the southwestern U.S. The semi-arid conditions cause the water table to be deep below the surface, implying that ground water does not play a large role in spreading contaminants from the surface, though meteoric waters possibly could. At the Nopal I site the water table lies about 100 meters below the uranium-mineralized zone (Wong 1994). This hydrologic regime is also similar to that of Yucca Mountain, Nevada where the water table lies 167 meters below the level at which the waste is to be buried (Wong 1994; www.ocrwm.doe.gov/documents).

3. DESCRIPTION OF THE SOILS

The main focus of this study has been the soils surrounding boulders of high-grade ore from the Prior High Grade Stockpile. The site where these samples were collected lays on a dipping slope away from the Nopal I mine. The slope is estimated to be about 11º. Even though a systematic description has not been done, a simplified one will be done here. Samples were taken from several different levels into the subsurface. All samples had pebble to clay-sized grains and were dark chocolate in color. Most of the large grains tend to be angular suggesting that they have not traveled far and are composed of the local rocks. The pebble-sized grains tended to be weathered pieces of tuff, while smaller factions may be tuffaceous or have an alternate origin. This area experiences high winds that could bring in other material. All samples also contained some organic fraction. This can be roots, small leaves, or twigs. Some body casts from insects have also been found.

4. PREVIOUS STUDIES IN RADIONUCLIDE MOBILITY AT NOPAL I

Radionuclide mobility at the Nopal 1 deposit has been studied in the past. Southwest Research Institute (SWRI) did some of the first work in the 1990s (Pearcy et al. 1995; Prikryl et al. 1997). Their main goal was to describe the petrology of the brecciated zone and uranium source and, to monitor migration of uranium radionuclides in the E-W fracture zone. In these studies, gamma-ray spectroscopy and other techniques were employed. They discovered secular disequilibrium in the fracture infillings. Ratios greater than unity were found when comparing $^{234}U/^{238}U$, $^{230}Th/^{234}U$, and $^{226}Ra/^{230}Th$ inside and outside the brecciated zone of the Nopal 1 deposit. They determined that there were excesses of ^{234}U, ^{230}Th, and ^{226}Ra in the fracture infillings outside the brecciated zone. Their interpretation was multistage mobilization that requires $^{230}Th > ^{234}U > ^{238}U$. Based on the half-life of ^{234}U they placed mobilization of the radionuclides within the last million years.

Wong and others conducted a similar study in 1999 (Wong et al. 1999). Again, gamma-ray spectroscopy was employed, and the fracture infillings and ore were analyzed. They found secular disequilibrium between ^{230}Th and ^{234}U and also between ^{226}Ra and ^{230}Th similar to SWRI. Inside the brecciated zone, daughter/parent (D/P) ratios of $^{230}Th/^{234}U$ and $^{226}Ra/^{230}Th$ were greater than unity. They also believed that the greater than unity ratio for $^{226}Ra/^{230}Th$ outside the breccia zone means precipitation rather than leaching. The most pronounced mobility was in veins and fractures with oxidized alteration minerals, e.g. hematite. Contrary to the studies at SWRI, they thought that mobility had been within the last 8 ka, based on the mobility of ^{226}Ra.

A recent study of the Nopal 1 deposit was conducted by Murrell and others in 2002 (Murrell et al. 2002). Thermal Ion Mass Spectrometry (TIMS) was used to analyze samples from