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AUTHOR(S) J. L. Thompson

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Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

RADIONUCLIDE MIGRATION STUDIES AT THE NEVADA TEST SITE

J. L. THOMPSON

LOS ALAMOS NATIONAL LABORATORY, MS-J514

LOS ALAMOS, NEW MEXICO 87544

Radionuclide Migration

Abstract

The United States government routinely tests nuclear devices at the Nevada Test Site (NTS) in southern Nevada. A significant amount of radioactive material exists underground at the NTS with no containers or engineered barriers to inhibit its subsequent migration. The Department of Energy has sponsored for many years a research program on radionuclide movement in the geologic media at this location. Goals of this research program are to measure the extent of movement of radionuclides away from underground explosion sites and to determine the mechanisms by which such movement occurs. This program has acquired significance in another aspect of nuclear waste management because of the Yucca Mountain Project. Yucca Mountain at the NTS is being intensively studied as the possible site for a mined repository for high level nuclear waste. The NTS provides a unique setting for field studies concerning radionuclide migration; there is the potential for greatly increasing our knowledge of the behavior of radioactive materials in volcanogenic media. This review summarizes some of the significant findings made under this research program at the NTS and identifies reports in which the details of the research may be found.

1. Introduction

In 1973 the manager of the US Department of Energy Nevada Operations Office established what became known as the Radionuclide Migration project. The goal of this project was to quantify the movement of radionuclides away from nuclear explosion zones and to define the factors that contribute to this movement. Responsibility for management of the program was assigned to technical co-directors from Los Alamos National Laboratory (LANL) and Livermore National Laboratory (LLNL) and a manager from the Nevada Operations Office. Other organizations participating in this project included the US Geological Survey (USGS) and the Desert Research Institute (DRI), as well as various support groups at the NTS. In addition to field determinations of radionuclide migration, these organizations participated in collecting and compiling existing data and in laboratory studies of radionuclide interactions with geologic materials from the NTS. The initial emphasis of this work was on near-field movement; long-range migration was part of area hydrology studies. In recent years the program has expanded to include hydrology studies on and adjacent to the NTS, and the program is now called the Hydrology/Radionuclide Migration Program (HRMP). The Nevada Operations Office takes an active role in managing the

program with additional assistance from an independent advisory committee of technical experts. HRMP research activities now include studies of most aspects of the environmental impact due to weapons testing. The major effort, however, remains the same: to understand those factors that affect the underground movement of radionuclides in the NTS environment. Although some purely hydrological studies are conducted by the HRMP, this review will focus largely on that part of our work which involves radionuclides.

The first major work undertaken by the HRMP was to compile a comprehensive review of what was known about the migration of radionuclides in groundwater at the NTS. This report [1] contains a description of the geology and hydrology of the NTS, a tabulation of the amount of radioactivity deposited underground as a result of testing through mid-1975 and a proposed program for laboratory and field work to expand the data base. The first field study was begun in 1974 at the site of the Cambric test which had been conducted in 1965. The cavity was at a depth of 294 m in alluvial soil and below the water table. Water movement from the cavity region was induced by pumping from an adjacent well. After two years of pumping, the elution of radioactive materials at the pumped well was detected. The pumping has continued for 15 years and we now have a data base for tritium elution with which various theoretical models can be tested. A second field study was started in 1982 in the brecciated rhyolite of Pahute Mesa at the Cheshire site. Here there is a natural hydraulic gradient that may assist the movement of nuclides away from the cavity. The transmissive zones in the fractured rhyolite may allow observation of retardation and dispersion phenomena associated with fracture flow. The site of a third field study is in the layered tuffs below Yucca Flat, where radionuclide movement over surprisingly long distances occasionally occurs. Other field studies include measuring the effects of craters on infiltration of radioactive materials toward the water table and observing the movement of radioactive tracers vertically and horizontally from a ditch carrying the source materials.

The HRMP has acquired additional importance in recent years because of the possibility of a nuclear waste repository being located at Yucca Mountain on the western edge of the NTS. The Yucca Mountain Project (YMP) has expanded many of the laboratory experiments initiated by the HRMP. Both programs need data on radionuclide source terms and transportation. Because the YMP has not yet reached the stage in which extensive field work can be undertaken, the experience and information gained in HRMP research may prove to be especially valuable. Furthermore, the opportunity exists to conduct field testing at some locations on the NTS without the experimental constraints which necessarily apply at Yucca Mountain because of its unique status as a potential repository. This review is intended to: 1) present an overview of the work with radionuclides accomplished by the HRMP to date, 2) indicate our present research activities, and 3) point out the potential of the NTS as a study area for measuring the underground movement of radionuclides in volcanogenic media.

2. Field Studies

2.1. Cambric

The Cambric site is the location of a 0.75-kt nuclear test conducted in 1965 in tuffaceous alluvium. The cavity was 73 m below the water table; the cavity and chimney had refilled to the preshot water level before re-entry drilling in 1974. The nuclear explosion provided enough tritium to

act as a water tracer and yielded detectable levels of fission products, uranium and plutonium. On re-entry it was found that most of the radioactivity was still retained in the cavity region. During the drilling of the re-entry well (RNM-1), 67 sidewall core samples were taken; these were later analyzed for their radionuclide content and lithology. With the use of packers, water was selectively pumped from five separate zones around the cavity and analyzed for radioactivity. Finally, water was pumped from a satellite well (RNM-2S) 91 m away to induce a hydraulic gradient at the cavity. Researchers have continued to pump water from RNM-2S with only brief interruptions since 1974 and have continued to monitor the radionuclide content of the well effluent. A detailed account of the early stages of the work at Cambric is in Ref. [2]. Various aspects of the Cambric investigations are emphasized in several publications [3-5], and elution data are presented in annual reports issued by LANL [6-17]. The elution of tritium from the RNM-2S as a function of volume of water pumped is shown in Fig. 1. Similar elution data for ^{36}Cl and ^{85}Kr are shown in Fig. 2 and Fig. 3, respectively. Comparison of these figures reveals that the ^{36}Cl elution peak is well ahead of the peak for the tritium. This is due to anion exclusion [18]; these data are discussed in more detail in Ref. 11. The elution of the ^{85}Kr is slightly delayed relative to tritium, perhaps because of some retention by zeolites in the alluvium. As of September 1989, we have pumped about 88% of the source term tritium from the RNM-1 well. The tritium elution data accumulated over 15 years of pumping serves as a field experiment against which theoretical models may be tested. One model, based on Sauty's calculations for a two-dimensional, radial, converging flow field [19], is shown in Fig. 4. The fact that this model does not accurately predict the elution tail may be ascribed to time dependent dispersion [16,20], a phenomenon discussed by MATHERON and DE MARSILY [21]. Several other attempts to model the tritium elution have also been made [11,22]. The elution of ^{129}I at the Cambric site has been studied [12,13]. The iodine concentrations were measured by tandem accelerator mass spectrometry and neutron activation analysis. The elution peak for iodine seems much broader than for chlorine, but the data are so scattered that a detailed comparison is not justified. Besides the radionuclides already discussed, only one other has been positively identified in the effluent of the RNM-2S well. Traces of ^{106}Ru were reported [23] in samples collected in 1978-80; subsequent detection was not possible as this radionuclide has a half-life of 1.1 years. The chemical form of the ruthenium was not determined.

We routinely collect water from both the RNM-1 and RNM-2S wells and analyze these samples for ^{90}Sr and ^{137}Cs . The concentrations of these radionuclides has continued to decline as water is pumped away from the cavity region, but neither radionuclide has yet been detected in the discharge at RNM-2S. The absence of ^{90}Sr and ^{137}Cs at RNM-2S is reasonable because these cations are very strongly sorbed on volcanogenic rock [24]. Our measurements at Cambric seem to provide field confirmation of predictions based on laboratory experiments; that is, we observe the migration of neutral and negative species through the alluvium, but the movement of positive ions is much delayed.

The effluent from RNM-2S flows more than one km in an unlined ditch and discharges in Frenchman Flat. The ditch has been equipped with flumes to measure the flow and infiltration rates, and the surrounding vadose zone has been instrumented with lysimeters, tensiometers, neutron probes and resistance cells. Transport times from the ditch to sampling points along the recharge plume have been measured using tritium and bromide ions as tracers [25,26]. In general, it appears that vertical movement below the ditch is considerably more rapid than horizontal movement. We

Fig. 1. Tritium elution at the Cambric site.

Fig. 2. ^{36}Cl elution at the Cambric site.

Fig. 3. ^{85}Kr elution at the Cambric site.

Fig. 4. Elution of tritium at the Cambric site: model vs observed. (See Ref. 11 for details of model.)

anticipate conducting other experiments on radionuclide migration in the vadose zone at this site in the future.

2.2. Cheshire

The Cheshire device was fired in early 1976 in the brecciated rhyolite of Pahute Mesa at a depth of 1174 m, some 544 m below the water table. In late 1976 some water from the cavity region was obtained, but the pump became stuck in the hole (U20n) and sampling was not resumed until 1983. In 1983 and 1984 investigators collected water samples from depths of about 1200 m. Then in 1985 the bottom of the hole was packed off, the casing was perforated at 820 m, and more water samples were obtained. In 1987 the investigators drilled another hole (UE20n#1) 300 m from the Cheshire site. This hole was 1000 m in depth and penetrated several permeable zones thought to serve as pathways for water flow away from the cavity region. Water from both U20n and UE20n#1 is being sampled and analyzed for its radionuclide content. More details concerning our work at the Cheshire site are contained in reports from LANL [14-16] and LLNL [25,26]. In contrast with the Cambic site, there is a natural hydraulic gradient at Cheshire, and there are indications that some of the source term radionuclides have moved away from the cavity-chimney region with the flowing water. Concentrations of tritium, ^{85}Kr , ^{90}Sr , ^{125}Sb , and ^{137}Cs at the U20n hole have decreased with time, and these radionuclides are present in water samples from the auxiliary hole UE20n#1. It is particularly interesting that manganese, cobalt, cerium and europium isotopes (and even appreciable fractions of cesium) are observed to be in association with colloidal material [27], so this may be a field site for the study of colloid transport.

2.3. Yucca Flat

This part of the NTS has been heavily utilized for testing. Within this area investigators have drilled a number of holes in an attempt to better understand the movement of radionuclides in the layered tuffs underlying the region. Because this area is frequently used for nuclear testing, most of these holes were open for only a few months, and then were stemmed with cement. In 1984 tritium was found in water at an emplacement hole (U3kz); subsequent analyses revealed that fission products were also present. Researchers then embarked on a very detailed elemental and isotopic analysis of samples to identify the source of this radioactive material. By matching the relative amounts of rhodium 101 and 102 isotopes in U3kz samples with those from surrounding test sites, they eventually were able to assert with reasonable confidence that the source was a test called Sandreef conducted some 350 m to the north in 1977 [28]. A hole (UE3e#1) drilled 61 m north of U3kz encountered the same suite of radioactive materials. Gamma logging indicated that the radioactivity was confined to a narrow band at 658 m depth. Cobalt, ^{244}Cm and NaCl were added to U3kz before the test was conducted there. The neutron activation products ^{60}Co and ^{36}Cl , along with the ^{244}Cm , serve as unique tracers for the test, which was called Aleman. More details concerning the investigations conducted at this site are contained in reports from three meetings of a committee which considered the implications of radionuclide migration there [29]. After the Aleman shot was fired in U3kz in September of 1986, two more holes were drilled in the same vicinity. Ue3e#2 produced core from the same depth as UE3e#1, but without radioactivity, although some tritium was found in the water during drilling. A second attempt to obtain core bearing radionuclides was frustrated when hole UE3e#3 collapsed before coring at

depth could be initiated. We anticipate another drilling effort at this site in FY 1990.

Another study has been conducted somewhat north of the Aleman site. There the main interest was in the pressurized aquifer that had been encountered at depth in several locations in the Yucca Flat area of the NTS. A few months after the Gascon test, investigators drilled a hole (UE4t) adjacent to the site of the test but found no radioactivity from that test or from any other nearby test sites. The pressurized aquifer was encountered, however, and hydrology tests were conducted in this hole [30]. We do not yet know if there is a relationship between the pressurized aquifer and the movement of radionuclides within the geosystem.

Movement of radioactive materials over distances of a few hundreds of meters at the NTS has been noted in a very few reports and memoranda [31,32,33]. In at least one of these cases [31] fission products seem to have been injected into a fracture. A significant problem in our research at the NTS is the need to distinguish between movement of radioactive material caused by the explosion (e.g., fracture injection) and subsequent movement by groundwater transport. Considering that over 500 tests have been conducted underground at the NTS [34], there are remarkably few sites at which radioactive contamination of the groundwater or rock has been detected outside the immediate region of the explosion zone. Satellite wells drilled in the vicinity of the test sites Nash, Bourbon and Bilby have produced water samples with minimal amounts of radioactive materials [25]. As stated above, we caused some movement of fission products at the Cambic site by forced pumping, and have observed movement at the Cheshire site with natural groundwater flow. The Aleman site (U3kz, UE3e#1) provides us with yet another opportunity to study the movement of radioactive material away from an explosion zone.

3. HRMP Research Activities

The goal of the HRMP research is to qualitatively and quantitatively evaluate the movement of radioactive material away from individual sites of nuclear tests, and away from the NTS as a whole. We have organized our efforts into four work elements: source term, transport, local hydrology and regional hydrology. The hydrology studies are largely the responsibility of the USGS and DRI and will not be discussed in this review, except as they impact directly on the first two work elements.

3.1. Source Term

The source term of hazardous material at the site of a nuclear test may include a chemical source term, a radionuclide source term, and a hydrologic source term. The first two refer to the total inventory of materials (hazardous chemicals and radionuclides) present at the site, while the third refers to that portion of the inventory which is available for migration. In order to evaluate the potential for migration from a particular test site we need accurate information on the source terms for that site; cumulative data are required to evaluate possible migration on a regional scale, as from the whole of the NTS.

At a particular site on the NTS we generally are able to obtain (classified) data on the total amounts of fission products and fissionable material present after the test, so the radionuclide source term can be known. To date, very few data have been collected concerning the kinds

and amounts of hazardous chemicals associated with nuclear tests, so the chemical source term is largely unknown. We are presently beginning to collect the necessary data to learn if there is a significant source of hazardous chemicals associated with underground nuclear tests. The hydrologic source term is hard to predict on the basis of the other two source terms because much of the potentially hazardous material (radioactive or hazardous chemicals) is bound up in the form of melt glass or strongly sorbed on rock surfaces and is not available for migration. There is often some differentiation of fission products following the explosion, with volatile elements like iodine and antimony more likely to be found in the chimney above the crater and refractory elements like zirconium more likely to end up in the bottom of the cavity in the melt glass [35]. As the volatile elements are often more readily dissolved by inflowing groundwater, they will generally contribute more heavily to the radiologic source term than will the refractory elements. We are currently collecting data from postshot drillback debris on the spatial distribution of fission products in cavity-chimney complexes; we are also measuring the relative leach rates of these materials by groundwater. Our goal is to study enough current tests to discover what generalizations about the source term can be made and what parameters of the test (yield, rock type, degree of water saturation, etc.) influence the source term.

3.2 Transport

The term transport, as used in HRMP research, includes movement of material by fracture injection (as by a steam or water pulse), movement as colloidal particulates, movement with episodic water flow, and movement in solution with flowing groundwater. In order to investigate transport phenomena we need information on the hydrologic source term, and on the geochemistry, hydrology and geology of the underground environment. As our understanding of transport processes at the NTS increases, we will develop models to predict the movement of hazardous materials and radiochemicals under given sets of conditions. To date, most of our work deals with description of transport rather than prediction. Many of our data come from the Cambric site which was discussed earlier in this paper. Transport in the alluvium at Cambric has been confined to neutral or anionic dissolved species moving with flowing groundwater. We have measured the colloid content of water at the Cambric site; it is relatively free of colloids compared to water elsewhere on the NTS, and colloid transport is not a likely means of movement. The homogeneity of the alluvium and the absence of large blast effects from the low-yield test probably account for the apparent absence of fracture injection at the Cambric site. Even in this relatively simple system a transport model based on a conventional two-dimensional convection-diffusion equation does not adequately describe the elution of tritium (refer to Fig. 4 and the accompanying discussion). The Cambric site provides us with a substantial body of data with which we may validate various models proposed for the transport of dissolved species.

Information about transport at the Cheshire site is less certain because of the natural hydraulic gradients at this site and because the hydrologic source term was never measured definitively. The data do indicate movement of neutral and anionic dissolved species (tritium, krypton, antimony), but also of species which are probably cationic (strontium, cesium, cobalt, europium, potassium). Transport in the form of colloids is indicated for some of these radioactive elements [27]. We are continuing to monitor the changing concentrations of radionuclides in samples from the U20n and UE20n#1 holes at the Cheshire site.

The transport of radioactive material observed at the Aleman site on Yucca Flat seems to be confined to a fracture. We expect to measure the depth of diffusion of the radionuclides into the bulk rock in order to estimate the time the radionuclides have been in contact with the rock. This measurement should enable us to distinguish between movement caused by the test event (fracture injection) and movement which occurred subsequently with the relatively slower moving groundwater. Such an analysis is dependent on securing core material containing the fracture, and we have not succeeded in doing this to date. We are continuing to search for other instances on the NTS where long-range migration of radionuclides may have occurred.

We have initiated several studies of radionuclide movement through the vadose zone above the water table. Here there may be two-phase movement and movement due to episodic water pulses caused by rainfall. The latter case may be especially important at the NTS where collapse craters can serve as catchment basins which funnel precipitation through the underlying chimney and cavity regions [37]. The effluent from the pumped well (RNM-2S) at Cambric is a continuous source of traced water which we may use to evaluate the movement of water vertically and horizontally from the discharge ditch. The instrumentation described above in section 2.3 has been used to determine that about 75 days are required for bromide ion added in the ditch to penetrate to a depth of 30 m [26]. Similar experiments with both radioactive and non-radioactive tracers are planned for the future.

4. The NTS as a Study Site

The NTS is a unique environment for the conduct of field studies of radionuclide migration underground. It is remote, hence transportation expenses for equipment and personnel may be considerable, and there are security requirements for personnel working on the site. But there are many advantages of this site, also. Numerous source terms are already in place both in the vadose and saturated zones; source terms with ages ranging from months to over 20 years are present. Typically, source terms include a variety of chemical species allowing observation of conservative and nonconservative tracer migration. Also there are several sites (U3kz, U4t) in which tracers were added before the underground explosion so that materials from those tests can be uniquely identified. There are sites at which transport of radionuclides under the impetus of episodic or continuous moisture pulses through unsaturated materials may be observed. There are several sites at which "anomalous" long-range migration of radionuclides has occurred. And there is one continuously pumped well with 15 years of elution data that can be used for numerical model validation. Field data obtained in the course of HRMP research should be very useful to those who are evaluating the possible establishment of a nuclear waste storage facility at Yucca Mountain.

The HRMP seeks to take advantage of the potential of the NTS for research purposes. We have studies underway at the NTS that should lead to a better understanding of radionuclide movement through alluvium (Cambric), fractured rhyolite (Cheshire), and layered tuff (Yucca Flat). We also are conducting studies of groundwater movement and seeking to understand the effect of nuclear testing on the local hydrology. The experience and information gained to date in the HRMP provide a strong foundation on which a much larger program of field research could be built. The unique potential for such a research program at this location should not be overlooked.

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