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STUDIES OF FISSION PRODUCT MOVEMENT IN TUFFACE· US MEDIA

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Radionuclide migration

Abstract

For approximately 25 years the United States has conducted underground nuclear tests at a site in the state of Nevada. These tests have left a variety of fission products at depths of 100 to 1000 meters below the land surface. The geologic media here consist primarily of tuffs and rhyolites. More than 150 tests were conducted at or below the water table. We are studying locations of past tests to determine whether residual fission products move through the underground environment and, if so, by what mechanisms. Our research involves consideration of leaching, sorption, hydraulic dispersion, fracture flow and colloid transport. The data we obtain are relevant to groundwater contamination and nuclear waste storage issues. In this paper we present information obtained from our research at several different locations within the study site. Specifically, we describe the movement of radionuclides including tritium, ^{85}Kr, ^{90}Sr, ^{106}Ru, ^{125}Sb, and ^{137}Cs in situations where groundwater was moving and in which it was relatively static.

1. Introduction

The information contained in this paper came from research aimed at discovering how radioactive materials move in saturated or partially saturated tuffaceous rock. Some of this research was carried out in the laboratory, but most of the data reported here are from field studies. The information which has been obtained is important for the US Department of Energy in this agency's attempts to assess environmental damage which may have occurred at its facilities and to rectify such damage. The information is also useful in efforts being made at the Nevada Test Site (NTS) by both federal and state agencies to protect the groundwater from adverse effects associated with underground nuclear testing. Finally, what we are learning about underground movement of radioactive and other hazardous materials may be relevant to the possible placement of a nuclear waste storage facility at Yucca Mountain on the western edge of the NTS.

Our field work was conducted on the 3600 km^2 NTS located in semi-arid southern Nevada where underground nuclear testing has occurred since

1962. Most tests were in tertiary rhyolites and tuffs, through a few were in tuffaceous alluvium. The water table at the NTS is generally 150 to 750 m below the land surface; the hydraulic conductivity is in the range of 1- to 100- m/y. Nearly one third of the more than six hundred underground tests conducted at the NTS were below the water table, and the cavities produced by these tests have since refilled with water.

In discussing the behavior of radioactive materials in the underground environment at the NTS, it is important to remember that they were a product of a nuclear explosion. Rock, device construction materials, stemming materials, electrical cables, everything in the immediate vicinity of the device is initially vaporized; the major fraction of radioactive material ends up in the "melt glass" produced when this vapor cools to a liquid and then a solid. Some radionuclides remain in the gas state (e.g., ^{85}Kr), some are condensed on the surface of the cavity walls. If water flows back into the cavity, some radionuclides will dissolve. The enormous pressures generated at the time of the explosion may cause radioactive materials to be injected into fractures; in a few instances fracture injected material has been found hundreds of meters from the cavity. Moving groundwater may carry dissolved species, and perhaps colloids, away from the initial site of deposition. A more complete description of the NTS and of the processes which accompany an underground nuclear explosion is contained in Ref. [1].

The research program concerning movement of radionuclides in the underground environment of the NTS was started in 1973 and the first field experiment began the following year. The program involves a number of organizations, including Los Alamos National Laboratory, Lawrence Livermore National Laboratory, the US Geological Survey, the Desert Research Institute, the Nevada Operations Office of the US Department of Energy, and various support companies operating at the NTS. The data acquired by this research program is now being used by a number of agencies concerned with groundwater protection, environmental monitoring, environmental remediation, and regulatory compliance with federal and state laws. Ref. [2] gives a more complete history and description of the scope of this program. The discussion which follows concerns two study sites at the NTS. These sites are in different geologic and hydrologic environments and the research methodology being employed is different. It is hoped that the description of our on-going research at these two sites will convey a sense of the problems and opportunities presented by the sources emplaced at the NTS, as well as describe what we have learned about how certain radionuclides move underground through tuffaceous media.

2. The Cambric Site

In 1974 we drilled into the cavity of the nuclear test Cambric which had been fired in 1965. This 0.75-kt test was in tuffaceous alluvium below the water level; there is almost no hydraulic gradient at this location. On reentry we found that the fission products and tritium associated with this test had remained in the cavity region. Analysis of cores and water samples enabled us to measure the distribution of ^{90}Sr, ^{106}Ru, ^{125}Sb, ^{137}Cs, ^{144}Ce, ^{147}Pm, and ^{239}Pu between rock and water in the cavity. Tritium and ^{85}Kr were measured in the water. Also in 1974 we began pumping water from a satellite well 91 m from the cavity. Pumping from this well has continued since this time and we have monitored the effluent to determine which dissolved radionuclides are transported with the groundwater under the conditions at this site. We also periodically measure the concentrations of radionuclides in water samples from the cavity region. The early work at Cambric is described in Ref. [3]; data are reported annually in a series of Los Alamos reports of which Ref. [4] is the most recent.

We may summarize our observations at the Cambric site as follows:

A. The tritium and fission products associated with the nuclear test remained in the cavity region for ten years. There was no moving groundwater to transport dissolved species; the energy produced by the explosion was too small to cause extensive fracturing and injection of material into the fractures. The distribution of radioactive elements between rock and water was consistent with distribution coefficients measured in the laboratory [5,6].

B. When an artificially induced hydraulic gradient was imposed, dissolved radionuclides were transported. After pumping over 1.6×10^7 m^3 of water from the satellite well more than 90% of the tritium originally in the cavity has been removed. We have observed the transport of ^{85}Kr, ^{36}Cl, ^{129}I, ^{106}Ru, and ^{99}Tc. The ^{85}Kr is dissolved as a neutral species; all the others are anionic. Anion exclusion caused the chloride ions to elute slightly ahead of the tritium [7]. We have not yet detected cationic species such as ^{90}Sr or ^{137}Cs in the effluent from the satellite well, but the concentrations of these radionuclides in the cavity has decreased appreciably during the years of pumping. The concentrations in the cavity of ^{137}Cs and ^{90}Sr as a function of volume of water pumped from the satellite well are shown in Fig. 1. Presumably, ions like Cs^+ and Sr^{++} are sorbed on the surfaces of the rock matrix through which the groundwater is moving. When the pumping experiment is over, it is our intention to sample the region between the two wells to determine how far those radioactive species with high sorption coefficients moved.

C. The experiment at Cambric can be viewed as a traced well-to-well pump test, and our 17 years of data provide a reference set against which various models may be tested to see how well they describe water flow under these circumstances. Sauty's two-dimensional, radial, converging flow field model [8] has been applied to these data [9]. Fig. 2. shows that the agreement between model and experiment is quite good over most of the range of data; time dependent dispersion may account for the lack of congruency in the curves for the later stages of pumping [10].

D. The effluent water from the satellite well flows in an unlined ditch for about a kilometer; this water, with known concentration of tritium, is used for infiltration studies. The vadose zone in the vicinity of the ditch has been instrumented with lysimeters, tensiometers, neutron probes and resistance cells in order to measure vertical and horizontal water profiles [11].

3. The Yucca Flat Site

At the Cambric site we were able to characterize a source of radionuclides, then see what changes occurred when groundwater was pumped from the vicinity of that source. In contrast, at our Yucca Flat study site a source of radionuclides was discovered in an unexpected location, and our efforts have been focused on determining how these radionuclides got to the site where they were located. This research has required us to drill a number of holes (four to date) in the vicinity in order to obtain rock cores, sidewall samples, and water samples from various depths in the water table. We are not able to pump large volumes of water from any of these holes and must infer from other methods whether water played a significant role in the movement of the radioactive material at this site.

Yucca Flat consists of alluvial material overlying tuff -- air fall, ash flow and layered tuffs. Hydraulic conductivity is mainly by fracture flow. The water table is about 500 m below the land surface. In 1985 tritium and fission products were found in water collected from an emplacement hole in Yucca Flat. The fission products included ^{85}Kr, ^{102}Rh, ^{106}Ru, ^{125}Sb, and ^{137}Cs. A chemical separation and isotopic analysis of rhodium in material from the emplacement hole enabled us to identify the nuclear test which generated the fission products [12]. This test had taken place in 1977 about 350 m away. Since the emplacement hole contained a steel liner, it was not possible to determine exactly where the radioactivity was entering the hole.

Another hole, UE3e#1, was drilled next to the emplacement hole; a narrow zone at 659 m depth was identified as containing the same suite of fission products and tritium. Both UE3e#1 and the emplacement hole were lost to further investigation when a nuclear test was conducted there in 1986. In 1987 we drilled UE3e#2 about 12 m from UE3e#1 and cored through the depth where radioactivity had been found in the first hole. No radioactive material was found in the core, although water from this depth did contain tritium. We concluded that the activity had moved from its original source (the test in 1977) through a fracture, and the fracture was not horizontal and of large areal extent as might be expected along a bedding plane. An attempt to drill another hole, UE3e#3, at an angle through the depth of interest failed because of cave-ins.

In 1990 we drilled UE3e#4 a few meters from the site of UE3e#1. The history of this hole and a comprehensive description of the geological and radiological analyses associated with this site have been published elsewhere [13]. Sidewall cores from this hole contained tritium, ^{106}Ru, ^{125}Sb, and ^{137}Cs. Water samples contained tritium, ^{85}Kr, and ^{137}Cs. Gamma logging showed the radioactivity to be confined to two narrow zones at 679- and 658-m. We brought back to the laboratory several samples of the zeolitized tuff from the zones where radioactivity was encountered in this hole; some of these samples contained ^{137}Cs and some did not. We did a series of sorption experiments with this tuff to determine if it had properties which would allow fission products to move through it a distance of hundreds of meters in nine years. The details of these experiments are given elsewhere [14]; a brief description follows here.

In one experiment we sorbed ^{137}Cs at one end of a 5-cm long column and pumped groundwater through the column at a rate corresponding to a linear velocity of 62 m/yr (a rate probably greater than groundwater moves in Yucca Flat). The experiment was discontinued after 161 days as no ^{137}Cs had been eluted. The tuff was extruded from the column, quartered, and each quarter counted. The ^{137}Cs had remained for the most part in the quarter where it had been originally sorbed; very little migration along the column was indicated. In another experiment we loaded the column with tuff from UE3e#4 containing ^{137}Cs and pumped water through it at a velocity of about 30 m/yr. In 81 days no radioactivity was detected in the effluent and the experiment was discontinued. As a final check on the sorptive properties of this tuff with respect to cesium, we conducted a batch sorption experiment using our usual protocols [6] for such work. After 21 days of contacting ^{137}Cs solution with the tuff the phases were separated by centrifugation, and the distribution of the radionuclide between solid and liquid were measured. We measured the sorption ratio to be 4×10^3; this value is in the range expected for zeolitized tuffs from the NTS [6]. We concluded from these laboratory experiments that the tuff at UE3e#4 in which the fission products were found was not likely to allow dissolved cesium to move through it a distance of hundreds of meters in nine years. Furthermore, the absence of radioactive colloidal material in the effluent from the columns suggest that colloid transport was not the means by which fission products moved at UE3e#4.

The research site at Yucca Flat which we have described seems to illustrate a situation in which an underground nuclear explosion injects radioactive material through a fracture to a considerable distance away. The evidence that this occurred at our study site is circumstantial; we have no direct proof that movement of fission products occurred at the time of the explosion. However, the fact that the radioactive materials are confined to very limited zones and that the tuff is very sorptive of one of the most abundant fission products seems to argue against movement under the impetus of moving groundwater. Furthermore, at other sites we do have evidence for fracture injection [15]. The most compelling evidence is from a site at which ^{192}Ir as well as fission products were found in fractures in an emplacement hole 170 m from a nuclear test in which iridium was loaded as a tracer. These materials were found two months after the nuclear device was detonated; the fractures were well above the water table, so water transport was not a possibility. The study at the Yucca Flat site is continuing. We are monitoring radioactivity and hydrostatic pressure at three depths in hole UE3e#4. In an adjacent area we have drilled into a chimney/cavity to evaluate the distribution of radionuclides in the postshot environment.

4. Summary

The two study areas described above are the field sites we have studied most intensively for radionuclide migration at the NTS. One site demonstrates that certain radioactive elements can be mobilized by flowing groundwater; the other suggests that radioactivity may be dispersed from a nuclear test by means other than groundwater transport. There are many more nuclear test sites of varying ages and in different geologic and hydrologic environments at the NTS. There are probably few places in the world at which such a large number and variety of radioactive sources are in place underground. The US Department of Energy, as well as the State of Nevada, have strong incentives to evaluate the environmental hazard associated with these sources. We believe that the NTS provides a very significant opportunity for research on radionuclide migration.

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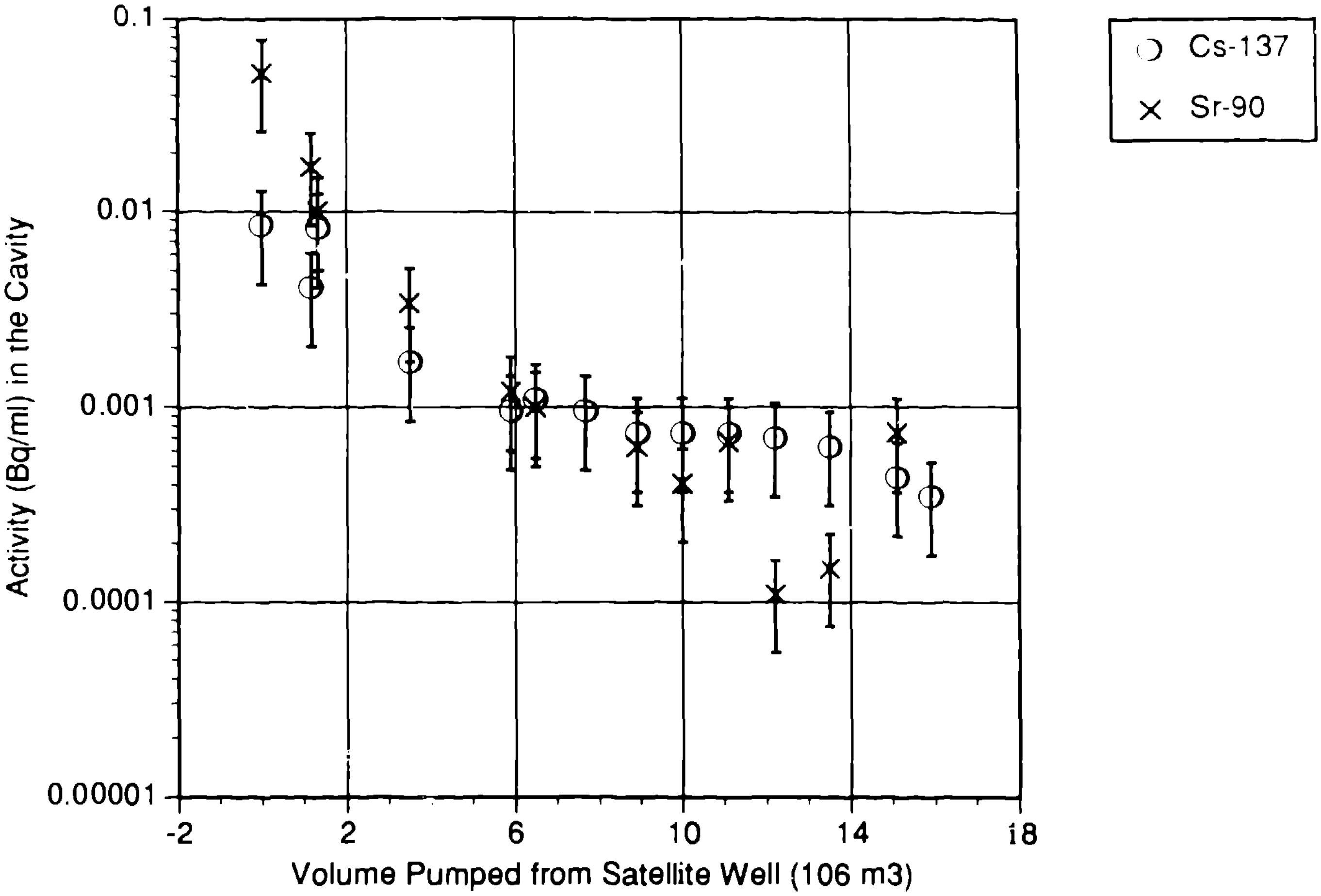
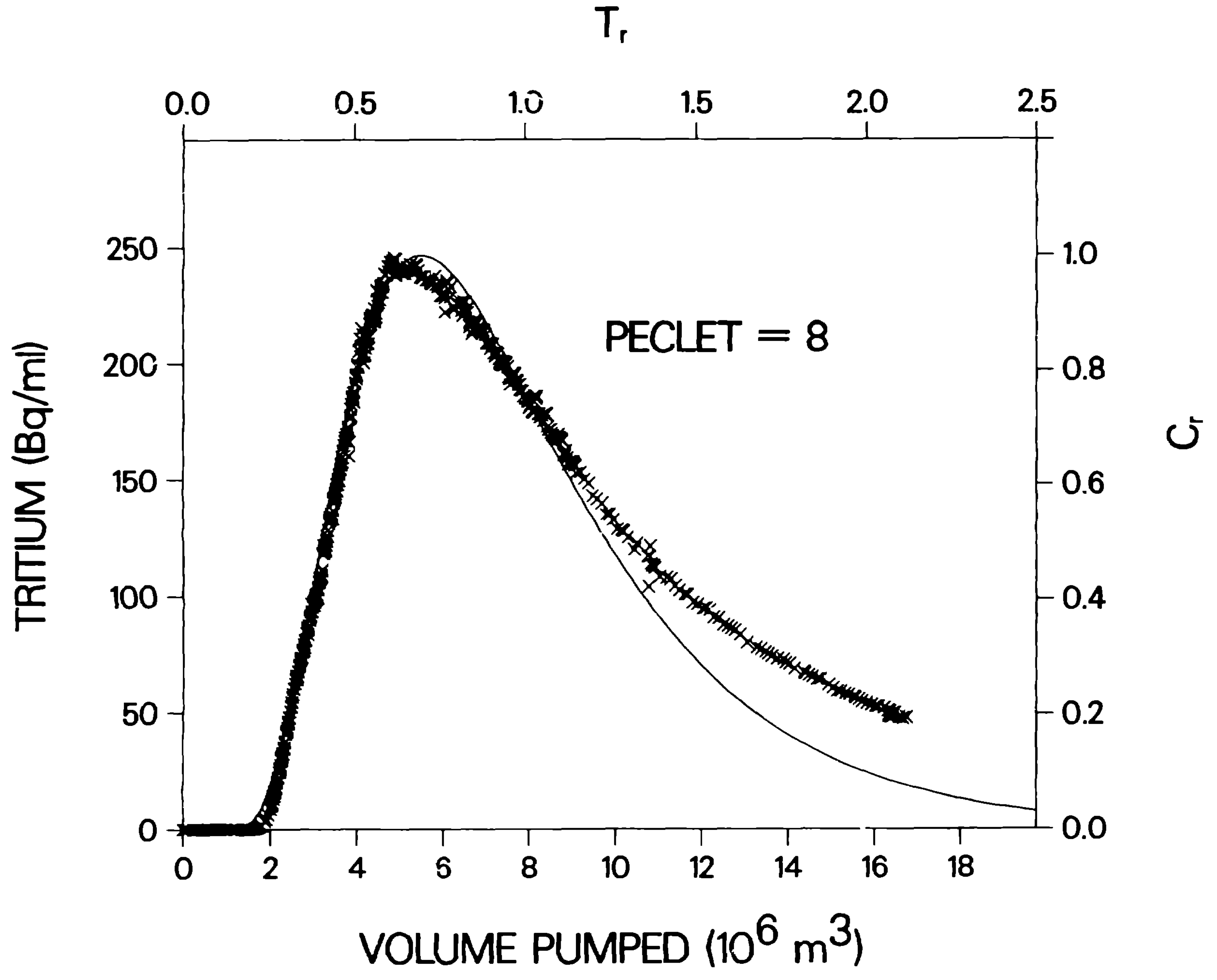


Cs-137
Sr-90
Activity (Bq/ml) in the Cavity
0.1
0.01
0.001
0.0001
0.00001
-2
2
6
10
14
i8
Volume Pumped from Satellite Well (106 m3)



T_r
0.0
0.5
1.0
1.5
2.0
2.5
250
200
150
100
50
0
TRITIUM (Bq/ml)
PECLET = 8
1.0
0.8
0.6
0.4
0.2
0.0
C_r
0
2
4
6
8
10
12
14
16
18
VOLUME PUMPED (10^6 m^3)