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Laboratory and Field Studies Related to the Radionuclide Migration Project

October 1, 1984—September 30, 1985

Compiled and Edited by Joseph L. Thompson

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ABSTRACT

In this report we review work performed in FY 1985 for the Radionuclide Migration program. Monitoring of water pumped from the satellite well at the Cambic site shows the continuing elution of tritium and krypton from the cavity but no appearance of fission-product cations. Water samples taken from two different depths in the re-entry hole at Cheshire have rather similar concentrations of radionuclides. This result, along with anomalously low tritium concentrations, makes the interpretation of data from Cheshire somewhat uncertain. We have made significant progress in laboratory studies of radionuclide sorption, actinide speciation, and colloid detection and migration.

I. INTRODUCTION

The Radionuclide Migration (RNM) project is an interagency program initiated in 1973; its goal is to determine the extent to which radionuclides produced by underground nuclear tests move within or outside the boundaries of the Nevada Test Site (NTS). Participating organizations include 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 organizations at the NTS. Los Alamos National Laboratory's contributions to the RNM program are in radionuclide detection, geochemistry, and the study of radionuclide/rock interactions. We have two principal study areas at the NTS, one at Frenchman Flat in Area 5 and the other on Pahute Mesa in Area 20. At the Cambrie site in Area 5, a large-scale field study has been under way since 1974 and has provided a great deal of information concerning the movement of radioactive species through tuffaceous alluvium. The Cheshire site in Area 20 provides the opportunity to study radionuclide movement in fractured rhyolite. Although the levels of radioactivity at both of these sites are relatively low and are not hazardous from a health physics perspective, a wide variety of radioactive species are present in sufficient concentrations so that we can study their behavior in a natural underground environment with a hydraulic gradient. Thus we can corroborate predictions based on laboratory studies and validate mathematical models that describe radionuclide migration. There are obvious areas of mutual interest between the RNM program and the Nevada Nuclear Waste Storage Investigations (NNWSI) project, and Los Alamos personnel contribute to both programs.

In this report we will review the work accomplished in the RNM program at the Cambrie and Cheshire study sites and at several other sampling sites on the NTS. We will also identify studies conducted at the Laboratory that have a direct bearing on RNM programmatic interests.

II. CAMBRIC STUDIES

In May 1965, the nuclear test Cambrie was conducted at Frenchman Flat on the NTS. The detonation point was 73 m below the water table (and 294 m below the ground

surface) in tuffaceous alluvium; disruption of the site caused by the 0.75-kt yield was relatively small. On re-entry of the Cambria cavity 9 years after the test, we found that the tritium and fission products had remained in the cavity and chimney regions, and we were able to measure the distribution of radionuclides between the water and soil. To determine how the radioactive material would respond to water movement, we created an artificial hydraulic gradient by pumping water from a satellite well 91 m from the cavity. This well has been pumped continuously since 1975; there has been frequent sampling of the effluent water pumped from the RNM-2S satellite well and occasional sampling of the cavity water *via* the re-entry RNM-1 hole. Our work at the Cambria site has been described in a number of articles¹⁻³ and annual reports.⁴⁻⁸ In this report we present data accumulated during the past year and discuss how these data are related to information acquired in previous years.

A. Sampling and Counting Methodologies

The quantities of radionuclides present in the water at the RNM site are quite small in many instances; this has necessitated rather large sample volumes and long counting times. In this section we will describe the techniques we employ to collect and measure water samples from the RNM site and other similar locations at the NTS.

Water samples for tritium analyses are usually acquired by pumping the water to the surface and collecting it in plastic bottles or jugs. Aliquots of 1 to 10 mL are taken for liquid scintillation counting; Insta-Gel (Packard Instrument Company) is used as the scintillation cocktail. Routine samples and background and tritium standards are counted repetitively for 50-min periods. This technique allows us to measure activities down to background levels (about 5 dpm/mL) with good precision, so our normal detection limits are approximately 2 pCi/mL or 5×10^7 atoms of tritium/mL of sample. We can count samples with lower concentrations of tritium, but that requires longer counting periods to achieve good precision.

Water samples to be analyzed for ^{85}Kr are collected in evacuated 2-ℓ stainless steel tubes that are filled from hose bibs for surface-level samples or lowered into the well for at-depth samples. The water is transferred from these “pressurized tubes” on a vacuum

line and purged with a krypton carrier gas; the gas is then purified and collected in a counting container with a thin titanium window. A propane gas flow proportional counter is loaded alternately with background samples, ^{85}Kr standards, and the samples being analyzed. The krypton samples are repetitively counted for 100-min counting intervals over a period of several weeks, and the decay curve is checked against that expected for ^{85}Kr . The detection levels for this counting regimen are similar to the background for the system, which is about 13 dpm/ml of gas or about 3×10^5 atoms of ^{85}Kr /ml of water. As in the case for tritium, lower concentrations of ^{85}Kr can be measured, but at the expense of longer counting times.

We routinely collect large-volume samples of water for analysis of fission products and for storage in archival collections. These samples are pumped into 55-gal. plastic-lined steel barrels that are used only once. We analyze for gamma-emitting radionuclides by concentrating the water in a glass still to a volume of several liters, evaporating the concentrate to dryness, and forming the residue into a cylindrical solid that is counted with a germanium detector and associated multichannel analyzer. A computer program translates gamma-ray intensities into atoms of a particular radionuclide and corrects to an appropriate "zero time" reference point. Corrections for detector efficiency and source-detector geometry are evaluated by a counting standard; the standard is made by doping a barrel of clean water (from Well 5B at the NTS) with a suite of NBS-certified gamma-emitting radionuclides and evaporating it to dryness. Background corrections are made from evaporated 55-gal. samples of Well 5B water that are free of detectable levels of fission products. The germanium detector is shielded to minimize gamma radiation from the environment as well as scattered radiation from the source; counting times usually include a 1000- and 4000-min count to optimize the activity levels present in samples from Cambria. Using this collection and counting system, we can detect ^{137}Cs at concentration levels of about 1×10^3 atoms/ml of sample.

We measure ^{90}Sr in 2-ℓ water samples by concentrating the sample and then precipitating strontium as the nitrate. After a series of chromate precipitations to remove any barium contamination, the strontium is converted to the carbonate and then counted. The ingrowth of the ^{90}Yt daughter is monitored, which both increases the sensitivity of

the method and allows a check for any spurious beta activity in the final precipitate. The sensitivity of this method is approximately 5×10^5 atoms of $^{90}\text{Sr}/\text{m}\ell$.

Our radiochemical procedure for ^{137}Cs involves purifying the sample and counting the cesium perchlorate precipitate. After the 2- ℓ sample is concentrated, the cesium is precipitated as the perchlorate and then dissolved and precipitated as a silicotungstate. We do several additional perchlorate precipitations interspersed with ferric hydroxide scavenger procedures to ensure the final purity of the product. The cesium, appropriate reagent blanks, and standard sources are gamma counted. The sensitivity of our radiochemical method for ^{137}Cs is about 1×10^6 atoms/ $\text{m}\ell$.

Plutonium in our samples is analyzed after they are radiochemically purified by alpha counting using a Frisch grid detector. The purification process involves oxidation with perchloric acid, extraction with HDEHP (di-2-ethylhexyl phosphoric acid), precipitation with lanthanum fluoride, sorption on an anion exchange resin, and drying of the final eluate on a platinum plate. The chemical yield through the purification steps is determined by adding a ^{236}Pu tracer to the initial 4- ℓ water sample. The method is so sensitive that about 1×10^6 atoms of ^{239}Pu can be detected in 1 $\text{m}\ell$ of sample.

In addition to the routine procedures just described, we also employ such analytical techniques as tandem-accelerator mass spectrometry, neutron activation analysis, and isotope dilution mass spectrometry to measure radionuclides like ^{36}Cl , ^{129}I , and ^{99}Tc that may be present at very low concentrations in our samples. We discuss the use of these techniques in previous reports^{5,6} and in Sec. II.D. of this report.

B. Tritium Migration (J. L. Thompson and S. Maestas)

During the past year the pump in the RNM-2S satellite well continued to operate at about 600 gal./min, and the tritium content of the pumped water continued to decline. The tritium concentration data for this year are tabulated in Table I, and complete elution curves are shown in Figs. 1 and 2. Through the end of September 1985, we had pumped about 3.9×10^4 Ci of tritium from RNM-2S, which is 65% of the original inventory. As the shape of the elution tail becomes better defined, we will be able to compare our model predictions^{6,8} with reality.

TABLE I
TRITIUM ANALYSES OF RNM-2S WATER SAMPLES

Identification Number	Date	Volume Pumped (10 ⁶ gals.)	Tritium ^a (10 ³ pCi/ml)
G 1437	10/18/84	2500.7	1.38
G 1438	11/16/84	2525.5	1.36
G 1439	12/21/84	2554.5	1.34
G 1440	01/25/85	2583.3	1.28
G 1441	02/21/85	2605.5	1.28
G 1442	03/18/85	2626.0	1.25
G 1443	04/19/85	2652.3	1.21
G 1444	05/21/85	2680.1	1.19
G 1445	06/03/85	2691.4	1.19
G 1446	07/01/85	2715.4	1.16
G 1447	08/16/85	2754.8	1.10
G 1448	09/04/85	2771.1	1.12

^aStandard deviation of counting data <1%.

C. Krypton Migration (J. L. Thompson, S. Maestas, and F. O. Lawrence)

In Table II we present the data from our monthly analyses of ⁸⁵Kr from water collected in the gas-tight pressurized tubes. The complete elution curve of krypton from RNM-2S is shown in Fig. 3. It is apparent that the maximum in this curve occurred at a volume of 6 to 7 x 10⁶ m³, which is significantly delayed relative to the maximum in the tritium elution curve. This suggests that the krypton is held up slightly during its passage through the alluvium, although in general it moves with the tritium. The atom ratio of krypton to tritium, shown in Fig. 4, continues to increase slightly but is still below the calculated source term ratio of 1.22 x 10⁻⁴.

Our annual pumping from the Cambrian cavity at RNM-1 yields samples that reflect the continued loss of tritium and krypton from this region. The tritium concentrations at RNM-1 are given in Table III, and in Table IV both tritium and krypton concentrations are shown with previously measured values. It appears that the tritium is now being removed from the cavity region more rapidly than is the krypton, perhaps because krypton

Fig. 1. Tritium concentration in water pumped from RNM-2S *vs* time. Values are corrected to Cambrian zero time.

Fig. 2. Tritium concentration in water pumped from RNM-2S *vs* volume pumped. Values are corrected to Cambrian zero time.

TABLE II
TRITIUM AND ^{85}Kr ANALYSES OF PRESSURIZED
WATER SAMPLES FROM RNM-2S^a

Identification Number	Date Collected	Volume Pumped (10^6 m^3)	Tritium Concentration		^{85}Kr Concentration		$^{85}\text{Kr}/\text{Tritium}$ Atom Ratio	
			(nCi/ml)	(10^{10} atoms/ml)	(dpm/ml)	(10^6 atoms/ml)	$\times 10^5$	Ratio ^b
434-1-84-016	10/04/84	9.423	4.07	8.47	0.749	6.12	7.22	0.59
434-1-84-018	11/02/84	9.515	4.02	8.37	0.678	5.54	6.62	0.54
434-1-84-020	12/06/84	9.625	3.91	8.14	0.535	4.37	5.37	0.44
434-1-85-001	01/04/85	9.716	3.97	8.27	0.670	5.47	6.62	0.54
434-1-85-004	02/15/85	9.847	3.86	8.03	0.643	5.25	6.54	0.54
434-1-85-005	03/01/85	9.890	3.84	7.99	0.649	5.29	6.62	0.54
434-1-85-010	05/21/85	10.146	3.66	7.61	0.606	4.94	6.49	0.53
434-1-85-011	06/03/85	10.188	3.70	7.70	0.596	4.87	6.32	0.52
434-1-85-013	07/01/85	10.279	3.60	7.49	0.563	4.62	6.17	0.51
434-1-85-014	07/01/85	10.279	3.63	7.55	0.632	5.17	6.85	0.56
434-1-85-016	08/16/85	10.428	3.55	7.38	0.488	3.97	5.38	0.44

^aAll activity levels corrected to Cambrian zero time (May 14, 1965).

^bMeasured ^{85}Kr /tritium atom ratio divided by ^{85}Kr /tritium atom ratio calculated for Cambrian zero time (1.22×10^{-4}).

Fig. 3. The ^{85}Kr concentration in water pumped from RNM-2S. Values are corrected to Cambrian zero time.

Fig. 4. Atom ratio of ^{85}Kr to tritium in water pumped from RNM-2S. Values are corrected to Cambrian zero time.

TABLE III
TRITIUM CONCENTRATION IN RNM-1 WATER (1985)

Identification Number	Volume Pumped (m ³)	Tritium Concentration (pCi/ml)	
		On Collection Date 4/10/85	Corrected to Cambic t _o 5/14/65
R-1	2.3	3.5	11
R-2	13.6	3.6	11
R-3	25.7	3.4	10
R-4	39.4	3.5	11
R-5	51.5	3.6	11
R-6	60.7	3.7	11
R-7	68.9	4.2	13
434-3-85-001 ^a	71.2	4.1	<u>13</u>
			Average 11

^aCollected in gas-tight pressurized tubes.

that initially sorbed on (or into) the alluvium now is being leached out. Note that the krypton/tritium atom ratio in the cavity region has remained well above the calculated source-term ratio.

D. Anion Migration (J. L. Thompson, J. F. Sledgianowski, and D. B. Curtis)

The nuclear test at Cambric produced relatively small quantities of ³⁶Cl and ¹²⁹I, and a portion of these radionuclides were dissolved in the groundwater at RNM-1. For example, we calculate⁸ that about 40% of the product ¹²⁹I was contained in the groundwater when we re-entered RNM-1 in 1974. Anions move through the alluvial soil at the Cambric site with minimal sorption, and both ³⁶Cl and ¹²⁹I have been detected in the water pumped from RNM-2S (Refs. 6 and 7).

Because of the very long periods required to obtain analyses of our ³⁶Cl samples by tandem-accelerator mass spectrometry, we are trying to develop alternative means of analysis. Liquid scintillation counting of ³⁶Cl is feasible if the samples can be sufficiently

TABLE IV
TRITIUM AND ^{85}Kr CONCENTRATIONS IN RNM-1 WATER

Zone	Date	Volume Pumped ^a (10^6 m^3)	Concentration ^b		$^{85}\text{Kr}/\text{Tritium}$	
			Tritium (10^9 atoms/ml)	^{85}Kr (10^6 atoms/ml)	Atom Ratio $\times 10^4$	Ratio ^c
Original-Zone IV	08/08/75	0.	3100.	570.	1.8	1.5
Original-Zone V	08/14/75	0.	790.	110.	1.4	1.2
Zone IV + V	10/04/77	1.17	66.	610.	92	75
Zone IV + V	11/30/77	1.34	41.	49.	12	10
Zone IV + V	09/04/79	3.50	5.4	4.6	8.5	7
Zone IV + V	10/05/81	5.89	3.0	1.5	5.0	4
Zone IV + V	04/01/82	6.48	1.6	0.55	3.3	3
Zone IV + V	01/06/83	7.67	0.70	0.24	3.4	3
Zone IV + V	04/24/84	8.91	0.46	0.19	4.1	3
Zone IV + V	04/10/85	10.0	0.27	0.21	7.8	6

^aTotal volume of water removed from RNM-2S by indicated date.

^bValues corrected to Cambrian zero time.

^cMeasured $^{85}\text{Kr}/\text{tritium}$ divided by $^{85}\text{Kr}/\text{tritium}$ calculated for Cambrian (1.22×10^{-4}).

concentrated and if interfering radionuclides are removed. We have developed a method of sample treatment involving precipitation of the chlorine with silver nitrate, filtration, and dissolution of the precipitate with sodium cyanide. Water samples of 10 to 20 ℓ from RNM-2S yield counting samples with activities several times background levels. The method is sufficiently promising that we will continue development using NBS standard ^{36}Cl .

We did no other analyses for anions during the past year but accomplished some preparative work that may eventually help us detect ^{99}Tc in RNM waters. We recently purified a ^{97}Tc source and calibrated it against an NBS ^{99}Tc standard. The ^{97}Tc was prepared by irradiating a ^{96}Ru target with neutrons to produce ^{97}Ru , dissolving the target, extracting the daughter ^{97}Tc , and finally purifying it by using anion exchange resins.

This ^{97}Tc can now be used in isotope dilution mass spectrometric analyses of ^{99}Tc in environmental samples.

E. Cation Migration (J. L. Thompson, S. Maestas, G. W. Knobeloch, J. Drake, D. W. Efur, and J. D. Gallagher)

One of the original goals of the Cambric experiment was to determine rates with which various radioactive species would move from the cavity at RNM-1 to the well at RNM-2S under the impetus of a hydraulic gradient. As noted above, we have traced the movement of neutral species (tritiated water, krypton) and anions (chlorine, iodine) during the 10 years we have pumped water from RNM-2S. To date, however, we have not detected in the RNM-2S any effluent species known to be cationic. We annually monitor the concentrations of the relatively long-lived fission products ^{90}Sr and ^{137}Ce at RNM-1, and although the concentrations have decreased with time, these radionuclides have not appeared at the satellite well. From the cumulative data in Table V it can be seen that the concentrations of ^{90}Sr and ^{137}Ce have not decreased in the cavity region as rapidly as has tritium. This probably indicates that some of the ions that were initially incorporated in the rock are now leaching out, giving relatively higher concentrations of these species. At some point in the future, a steady state may be achieved in which the leach rate and removal rate will be equal.

It is encouraging to note that the field test at RNM-1 confirms our predictions, which were based on laboratory studies: species with high sorption ratios are not readily transported through tuff by water. If ^{90}Sr or ^{137}Ce does appear at RNM-2S, it will be quite important to determine whether the radionuclide is in solution or has been transported in some other form (for example, as a colloid).

III. CHESHIRE STUDIES

The Cheshire test was conducted on Pahute Mesa at the NTS in February 1976. The cavity region was re-entered in 1976, but only a small amount of water was pumped out before 1983. The cavity location is well below the water table and in a region where there are natural hydraulic gradients, so there is the potential for movement of radionuclides

TABLE V
TRITIUM, ^{90}Sr , AND ^{137}Cs CONCENTRATIONS IN RNM-1 WATER

Zone	Date	Volume Pumped ^a (10 ⁶ m ³)	Concentration ^b			Atom Ratio x 10 ⁴	
			Tritium (10 ⁹ atoms/ml)	^{90}Sr (10 ⁶ atoms/ml)	^{137}Cs (10 ⁶ atoms/ml)	$^{90}\text{Sr}/\text{T}$	$^{137}\text{Cs}/\text{T}$
Original-Zone IV	08/08/75	0.	3100.	130.	23.	0.4	0.07
Original-Zone V	08/14/75	0.	790.	5.5	4.6	0.07	0.06
Zone IV + V	10/04/77	1.17	66.	22.	5.5	3.	0.8
Zone IV + V	11/30/77	1.34	41.	13.	11.	3.	3.
Zone IV + V	09/04/79	3.50	5.4	4.4	2.3	8.	4.
Zone IV + V	10/05/81	5.89	3.0	1.5	1.3 (1.6) ^c	5.	4 (5) ^c
Zone IV + V	04/01/82	6.48	1.6	1.3	1.5 (1.5) ^c	8.	9 (8) ^c
Zone IV + V	04/06/83	7.67	0.70		1.3 (1.1) ^c		19 (16) ^c
Zone IV + V	04/24/84	8.91	0.46	0.81	1.0	17.	23.
Zone IV + V	04/10/85	10.0	0.27	0.55	1.0	20.	37.

^aTotal volume of water removed from RNM-2S by indicated date.

^bValues corrected to Cambrian zero time.

^cValues in parentheses are based on gamma-spectral analyses of 55-gal. (evaporated) samples.

away from the explosion zone. The rock in this region is brecciated rhyolitic lava. We began studying the Cheshire site because it offered the opportunity to observe radionuclide migration under natural conditions and because the rock type was different from that being studied at the Cambria site. We extensively sampled the water pumped from Cheshire in September 1983 and again in August and October 1984. Routine monitoring of tritium took place between these major sampling efforts. The water pumped from Cheshire in 1983 and 1984 is supposed to have flowed into the re-entry hole through perforations in the casing at a depth of about 1200 m, close to the level of the cavity. Hydrologic studies in the region suggest that the natural direction of water movement is from the cavity to relatively permeable zones above, then laterally to the south and west. We wished to intercept water along this flow path, so in May 1985 the bottom of the hole was packed off; new perforations were made at a depth of approximately 820 m and pumping was resumed. In June 1985 we collected a number of samples of the pumped water for comparison with previous samples. In this report we present the results of three types of water analyses: (1) grab samples analyzed only for tritium, (2) pressurized samples analyzed for tritium and ^{85}Kr , and (3) large-volume samples analyzed for gamma-ray-emitting radionuclides and ^{90}Sr . The methodology employed in obtaining the sample and making the measurements was similar to that described in Sec. II.A. Results of our analytical work before October 1984 and more details concerning the earlier history of Cheshire are given in Ref. 8.

A. Radionuclides at 1200 and 820m (J. L. Thompson, S. Maestas, M. R. Cisneros, F. O. Lawrence, G. W. Knobeloch, D. W. Eford, J. Drake, and J. D. Gallagher)

Analytical data from Cheshire are presented in Tables VI-VIII. Although Table VI contains tritium concentration values obtained in FY 1985 only, Table VII includes ^{85}Kr and tritium measurements made in previous years as well. During the past year we rebuilt and recalibrated the counter system used for gamma-ray analysis of evaporated large-volume samples, so we recounted earlier samples as well as new ones. We have included in Table VIII all the measurements made with the rebuilt counting system. We also listed measurements of tritium, ^{85}Kr , ^{90}Sr , ^{137}Cs , and ^{239}Pu obtained by other techniques.

TABLE VI
TRITIUM CONCENTRATION IN WATER PUMPED FROM CHESHIRE

Identification Number	Date	Volume Pumped (10 ⁶ gals.)	Tritium 10 ² (nCi/ml) ^a
852-1-84-001	08/01/84	0.17	5.40
C-11	10/03/84	2.3	5.17
C-12	10/16/84	2.8	5.16
C-13	10/18/84	2.8	5.16
852-1-84-003	10/23/84	3.0	5.20
852-1-84-005	10/23/84	3.0	5.17
C-14	10/25/84	3.1	5.14
C-15	04/24/85	3.1	5.01
C-16	04/26/85	3.2	4.97
852-1-85-001	05/09/85	3.4	4.95
852-1-85-002	05/09/85	3.4	5.00
852-1-85-003	05/14/85	3.5	4.58
852-1-85-004	05/14/85	3.5	4.71
C-17	05/29/85	3.7	4.94
C-18	06/05/85	4.1	4.21
C-19	06/11/85	4.4	4.32
852-1-85-005	06/18/85	4.8	4.11
C-20	06/27/85	5.2	3.30
C-21	07/05/85	5.6	4.20
C-22	07/12/85	6.0	4.21
C-23	07/17/85	6.3	3.98
C-24	09/11/85	6.4	3.17

^aStandard deviation of counting data <1%. Values are corrected to $t_0 = 02/14/76$.

The tritium concentration in water pumped from Cheshire has diminished as water is removed from the hole. Data from Table VI are plotted in Fig. 5, and the approximately linear relationship between tritium concentration and volume pumped is apparent. It is particularly interesting that there is no change in the slope of the curve at the point (about 3.6×10^6 gals. pumped) when we began using the new perforations. This would seem to indicate that water drawn from the area adjacent to the cavity and water drawn from several hundred meters above the cavity contain the same amount of tritium—a very

TABLE VII
TRITIUM AND ^{85}Kr ANALYSES OF PRESSURIZED
WATER SAMPLES FROM CHESHIRE

Zone	Date	Volume Pumped (10^6 gals.)	Concentration		$^{85}\text{Kr}/\text{Tritium}$	
			^{85}Kr (10^9 atoms/ml)	Tritium (10^{12} atoms/ml)	Atom Ratio $\times 10^4$	Ratio ^a
852-1-83-008	09/09/83	0.06	5.5	12.0	4.6	0.78
852-1-83-009	09/09/83	0.06	5.9	11.6	5.1	0.86
852-1-84-001	08/01/84	0.17	5.7	11.2	5.1	0.86
852-1-84-003	10/23/84	3.0	5.3	10.8	4.9	0.83
852-1-84-005	10/23/84	3.0	4.4	10.7	4.1	0.70
852-1-85-001	05/09/85	3.4	5.6	10.3	5.4	0.91
852-1-85-002	05/09/85	3.4	5.4	10.4	5.2	0.88
852-1-85-003	05/14/85	3.5	5.5	9.5	5.7	0.96
852-1-85-004	05/14/85	3.5	5.7	9.8	5.8	0.99
852-1-85-005	06/18/85	4.8	4.8	8.5	5.6	0.95

^aMeasured $^{85}\text{Kr}/\text{tritium}$ divided by $^{85}\text{Kr}/\text{tritium}$ atom ratio calculated for Cheshire zero time (5.90×10^{-4}).

surprising result. Our data set for nuclides other than tritium is rather limited, but it is interesting to see how their concentrations vary as a function of volume pumped. Several such plots are included in Fig. 6. Antimony-125 and ^{85}Kr show the same behavior as tritium, but ^{137}Ce and ^{90}Sr in water pumped through the new perforations show a marked decrease in concentration. Most of the other radionuclides listed in Table VIII (many of which are at barely detectable concentrations) show relatively small concentration changes when the new perforations were put into use. The graph in Fig. 5 indicates another peculiar aspect of our data. There appears to be an approximate decrease of 40% of the initial tritium concentration during removal of 8×10^6 gals. of water, implying a total reservoir volume of about 20×10^7 gals. However, calculations based on the yield of the test device indicate a cavity nearly five times larger than this. We will discuss possible reasons for the apparently anomalous data from Cheshire in Sec. III.C.

TABLE VIII
RADIONUCLIDES IN WATER AT CHESHIRE^a

Date Collected	09/08/83	08/01/84	10/23/84	06/18/85	06/18/85
Ident. No. 852-9-	-83-006	-84-001	-84-003	-85-003	-85-004
	Volume Pumped (10 ⁶ gals.)				
	0.06	0.17	3.0	4.8	4.8
³ H	1.23 x 10 ¹³ (5.91 x 10 ⁻¹)	1.12x 10 ¹³ (5.38 x 10 ⁻¹)	1.08 x 10 ¹³ (5.19 x 10 ⁻¹)	8.5x 10 ¹² (4.1x 10 ⁻¹)	8.5 x 10 ¹² (4.1 x 10 ⁻¹)
²² Na	4 x 10 ⁴ (9 x 10 ⁻⁹)	3 x 10 ⁴ (7 x 10 ⁻⁹)	3 x 10 ⁴ (7 x 10 ⁻⁹)	8 x 10 ³ (2 x 10 ⁻⁹)	5 x 10 ³ (1 x 10 ⁻⁹)
⁴⁰ K	4.7 x 10 ¹² (5.1 x 10 ⁻⁹)	5.0 x 10 ¹² (5.4 x 10 ⁻⁹)	4.4 x 10 ¹² (4.7 x 10 ⁻⁹)	1.2 x 10 ¹³ (1.3 x 10 ⁻⁸)	8.3 x 10 ¹² (8.9 x 10 ⁻⁹)
⁶⁰ Co	2.3 x 10 ³ (2.6 x 10 ⁻¹⁰)	4.0 x 10 ³ (4.5 x 10 ⁻¹⁰)	1.2 x 10 ³ (1.4 x 10 ⁻¹⁰)	1.9 x 10 ³ (2.1 x 10 ⁻¹⁰)	8.3 x 10 ² (9.3 x 10 ⁻¹¹)
⁸⁵ Kr	5.7 x 10 ⁹ (3.2 x 10 ⁻⁴)	5.7 x 10 ⁹ (3.2 x 10 ⁻⁴)	4.9 x 10 ⁹ (2.7 x 10 ⁻⁴)	4.8 x 10 ⁹ (2.6 x 10 ⁻⁴)	4.8 x 10 ⁹ (2.6 x 10 ⁻⁴)
⁹⁰ Sr	7.0 x 10 ⁶ (1.5 x 10 ⁻⁷)	5.6 x 10 ⁶ (1.2 x 10 ⁻⁷)	5.3 x 10 ⁶ (1.1 x 10 ⁻⁷)	2.5 x 10 ⁵ (5.2 x 10 ⁻⁹)	
¹⁰⁶ Ru	2 x 10 ⁶ (1 x 10 ⁻⁶)	4 x 10 ⁶ (2 x 10 ⁻⁶)	2 x 10 ⁶ (1 x 10 ⁻⁶)	6 x 10 ⁶ (3 x 10 ⁻⁶)	2 x 10 ⁶ (1 x 10 ⁻⁶)
¹²⁵ Sb	1.5 x 10 ⁷ (3.2 x 10 ⁻⁶)	1.6 x 10 ⁷ (3.4 x 10 ⁻⁶)	1.3 x 10 ⁷ (2.8 x 10 ⁻⁶)	1.4 x 10 ⁷ (3.0 x 10 ⁻⁶)	1.5 x 10 ⁷ (3.2 x 10 ⁻⁶)
¹³⁴ Cs	3 x 10 ⁴ (1 x 10 ⁻⁸)	4 x 10 ⁴ (1 x 10 ⁻⁸)	4 x 10 ⁴ (1 x 10 ⁻⁸)	3 x 10 ³ (1 x 10 ⁻⁹)	Not detected
¹³⁷ Cs radiochem.	7.3 x 10 ⁷ (1.4 x 10 ⁻⁶)	1.2 x 10 ⁸ (2.4 x 10 ⁻⁶)	1.4 x 10 ⁸ (2.8 x 10 ⁻⁶)	3.6 x 10 ⁷ (7.1 x 10 ⁻⁷)	
¹³⁷ Cs gamma spec.	5.5 x 10 ⁷ (1.1 x 10 ⁻⁶)	6.3 x 10 ⁷ (1.2 x 10 ⁻⁶)	7.5 x 10 ⁷ (1.5 x 10 ⁻⁶)	3.1 x 10 ⁷ (6.1 x 10 ⁻⁷)	2.3 x 10 ⁷ (4.5 x 10 ⁻⁷)

TABLE VIII (cont)
RADIONUCLIDES IN WATER AT CHESHIRE^a

Date Collected	09/08/83	08/01/84	10/23/84	06/18/85	06/18/85
Ident. No. 852-9-	-83-006	-84-001	-84-003	-85-003	-85-004
Volume Pumped (10 ⁶ gals.)					
	0.06	0.17	3.0	4.8	4.8
¹⁵² Eu	10 ⁴ (10 ⁻¹⁰)	10 ⁴ (10 ⁻¹⁰)	10 ⁴ (10 ⁻¹⁰)	x 10 ⁴ (10 ⁻¹⁰)	10 ³ (10 ⁻¹⁰)
¹⁵⁴ Eu	1 x 10 ⁴ (7 x 10 ⁻¹⁰)	2 x 10 ⁴ (1 x 10 ⁻⁹)	8 x 10 ³ (5 x 10 ⁻¹⁰)	6 x 10 ³ (4 x 10 ⁻¹⁰)	7 x 10 ³ (5 x 10 ⁻¹⁰)
¹⁵⁵ Eu	10 ⁵ (10 ⁻⁸)	10 ⁴ (10 ⁻⁹)	10 ⁴ (10 ⁻⁹)	10 ⁵ (10 ⁻⁸)	10 ⁴ (10 ⁻⁹)
²³⁹ Pu	1 x 10 ⁷ (2 x 10 ⁻¹⁰)	4 x 10 ⁶ (1 x 10 ⁻¹⁰)	1 x 10 ⁷ (2 x 10 ⁻¹⁰)	4 x 10 ⁶ (1 x 10 ⁻¹⁰)	

^aUnits are atoms/mL and (μ Ci/mL), both corrected to $t_0 = 02/14/76$.

B. Comparison with Cambric

In previous reports dealing with the Cambric site (for example, Ref. 3), we calculated "retention factors," which are a measure of the extent to which radionuclides produced by the nuclear explosion are retained in the soil in either a fused or sorbed state. In our annual report for FY 1985 (Ref. 8), we noted that the rhyolite of Pahute Mesa seemed to have retention factors similar to those of the alluvium at Cambric. The radionuclide concentrations measured in Cheshire samples during this past year confirm these earlier observations, and it does appear that despite differences in both the geologic media and the strength of the nuclear explosions at the two sites, the retention of product radionuclides by the solid material is similar at Cheshire and at Cambric. These measurements are of some importance, because most of the nuclear testing now done at the NTS is in rhyolite or alluvium.

Fig. 5. Relative tritium concentration in water from Cheshire as a function of volume pumped. Values are corrected to Cheshire zero time.

Fig. 6. Relative radionuclide concentration in water from Cheshire as a function of volume pumped. Values are corrected to Cheshire zero time.

C. Data Interpretation

As indicated, there are still some uncertainties about the Cheshire study. In particular, we do not know the source of water that we are sampling. Four points must be considered. (1) The concentrations of nonsorbing radionuclides like tritium and ^{85}Kr appear to be the same at widely separated depths. (2) Fission products are present at both depths—even highly sorbing elements like strontium and cesium, which normally are not expected to move very far through geologic media. (3) The rate at which tritium diminishes with pumping implies a containment reservoir with a volume much smaller than the volume calculated for the cavity. (4) None of the concentrations of tritium sampled from Cheshire have approached the calculated source-term values⁸ for this radionuclide.

It may be possible that the water nominally drawn into the hole casing at the two different depths is in fact entering at only one point. Pressure tests of the hole indicated that it was tight before the second set of perforations were made, but after that test the casing may have cracked or the bridge plug may have begun to leak. The lack of great differences in the radionuclide content of the water before and after the second perforations were made argues that the waters come from the same or very similar sources. The presence of fission products suggests that the water has not traveled great distances from the cavity/chimney region. It may be possible that some fission products were instantaneously injected along a fracture at the time of the explosion and thus are located much further away from the cavity than is normally expected. If an appreciable fraction of the product radionuclides and tritium were contained in a fracture or offshoot of the main cavity in the vicinity of the re-entry hole, this might result in an apparently excessive rate of tritium removal when the hole is pumped. In any event, it is not clear what the spatial relationship is between the two perforations, our sampling point(s), and the cavity. We have two more large-volume samples to analyze, one taken just before the second perforations were made, and one taken in November 1985. It may be that the additional data provided by these samples will help resolve some of the uncertainties discussed above.

IV. STUDIES AT OTHER NTS SITES (J. L. Thompson, S. Maestas, and F. O. Lawrence)

Occasionally we have the opportunity to analyze water samples from sites that are not included in the regular RNM surveillance. Although such samples (for example, from emplacement holes) are usually available on a one-time-only basis, they can provide some information about the general status of radionuclide content in groundwater at the NTS. This past year we sampled water from two sites where emplacement holes had filled with formation water.

On April 9, 1985, we obtained three pressurized water samples from Hole U3kz. Our analyses indicated a tritium activity of $5.4 \times 10^{-2} \mu\text{Ci}/\text{m}\ell$ and a ^{85}Kr activity of $5.7 \times 10^{-5} \mu\text{Ci}/\text{m}\ell$ in this water. These activity levels are only a factor of 2 less than comparable values from Cheshire, so the water may have been drawn from the cavity region of a nearby earlier test. We hope to be able to obtain a large-volume sample from Hole U3kz to determine whether fission products are also present in the water.

In contrast with those from Hole U3kz, samples of water obtained from Hole U4t contained about $2.7 \times 10^{-7} \mu\text{Ci}/\text{m}\ell$ of tritium, which is essentially a background level. The Hole U4t sample was obtained September 11, 1985; we have no plans to analyze other samples from this hole.

V. LABORATORY AND COMPUTATIONAL WORK (B. P. Bayhurst, M. R. Cisneros, L. S. Hersman, D. E. Hobart, S. D. Knight, F. O. Lawrence, A. J. Mitchell, T. W. Newton, A. E. Ogard, M. A. Ott, P. D. Palmer, R. S. Rundberg, K. W. Thomas, J. L. Thompson, and B. J. Travis)

At this laboratory there is a long-term, extensive program pertaining to radionuclide interactions with the geologic media at the NTS. Initial research was conducted as part of the RNM project;¹ later, studies were supported jointly by the RNM and NNWSI projects.⁹ For the latter project, we are evaluating the potential of Yucca Mountain (on the western edge of the NTS) as a repository for nuclear wastes. A good deal of the NNWSI research is directly applicable to our RNM studies. For example, most of our understanding of sorption reactions of fission products on tuff has been gained through studies supported by

both projects.¹⁰ In this section we will review briefly some the NNWSI work accomplished during FY 1985 that is particularly relevant to the RNM project.

The effect of groundwater composition on radionuclide sorption has been a subject of study at Los Alamos for some time.¹⁰ We recently reported¹¹ that elements such as barium, cesium, and strontium, which sorb by an ion exchange mechanism, have appreciably lower sorption coefficients in a "high-salt" water such as that from Hole UE25p#1 than in a low-salt water such as that from Well J-13. Other elements, such as uranium and selenium, showed little variation in sorption coefficient between these two waters, presumably because they do not sorb by ion exchange. Water with relatively high pH and low Eh generally increased the tendency of dissolved radioactive species to sorb on tuff. Long-term studies indicate that technetium and neptunium form stable species in groundwaters, and their sorption behavior does not change over a 15-month period. This last information is particularly relevant for the RNM project because we anticipate measuring the technetium content of archival samples from RNM-2S.

The behavior of plutonium in a groundwater environment is not well understood, and several studies bearing on this subject are under way. We are attempting to measure saturation concentrations and equilibrium constants for the system Pu(VI)-Pu(V)-Pu(IV)-Pu(colloid), as well as the effect of radiolysis on these parameters.¹² Sorption or uptake of plutonium by microorganisms present in groundwaters is being evaluated;¹¹ this appears to significantly affect the amount of dissolved plutonium in the groundwater/tuff system. We reported recently¹² that plutonium tends to pass through short columns of crushed tuff at rates many times greater than would be predicted on the basis of sorption data. This behavior appears to be related to the oxidation state of the plutonium; Pu(VI) and Pu(V) are relatively mobile. Also, we see evidence that plutonium polymer and americium colloidal particulates pass through these columns.¹¹

The possibility of radionuclide transport in the form of colloids is receiving attention through a number of studies at the Laboratory. The concentration level of natural colloids present in water pumped from Well J-13 has recently been measured.^{11,12} We conclude that this water contains too little material to contribute appreciably to radionuclide transport of species that might sorb on the natural colloids. Our autocorrelation photon spectroscopy

system has been used to measure the size distribution of reference polystyrene beads in the range of 5 nm to 1 μ m and to measure the size distribution of plutonium polymers.¹¹ We have added population balance equations to the code TRACR3D so that we can simulate migration of colloids through geologic media.¹⁰ These studies of colloid transport will be of great value to the RNM program if we undertake a field study of colloid migration at the RNM-1 site.

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