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*Laboratory and Field Studies Related
to the Hydrology/Radionuclide
Migration Project*

October 1, 1988–September 30, 1989

Compiled and Edited by Joseph L. Thompson

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ABSTRACT

This annual report describes research conducted in FY1989 by Los Alamos National Laboratory in the Hydrology/Radionuclide Migration Project. The work, which is done in coordination with several other organizations, relates to the movement of radionuclides underground at the Nevada Test Site. This project continues the long-term monitoring of tritium and fission product concentrations in water at the Cambric study area. As water has been pumped from an adjacent well, radionuclide concentrations in the cavity region have diminished to near the limits of detection. Neutral and anionic radionuclides have been measured in the effluent of the pumped well, but cations such as ^{90}Sr and ^{137}Cs have not been detected. This year a study was initiated to measure the spatial distribution of refractory and volatile elements in the cavity/chimney regions of underground nuclear tests. Differences in both distribution and leachability were observed for the two sites studied. Colloidal material from the Cambric site was collected and analyzed by transmission electron microscopy. We believe we were able to distinguish natural colloids from those introduced in drilling fluids. We tested the hypothesis that the $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratio would be useful in making this distinction and concluded that it was not. We reported a first measurement of ^{99}Tc in water from the Cambric site. We also participated in the hydrologic study of a highly pressurized zone in the tuff aquifer at the Nevada Test Site that was apparently created by nuclear testing. Finally, this report includes a description of various program management and planning activities conducted at Los Alamos.

EXECUTIVE SUMMARY

This report describes the research activities conducted in FY1989 by Los Alamos National Laboratory (Los Alamos) personnel in the Hydrology/Radionuclide Migration Project. The goal of this project is to understand those factors that affect the movement of radionuclides (and other hazardous materials) underground at the Nevada Test Site (NTS). At Los Alamos, we have focused our research primarily on source terms and transport; we also contribute to hydrologic studies. Personnel involved in this work reside in several different Laboratory divisions. Other research programs, like the Yucca Mountain Project, study similar environmental issues.

Program management of this project includes normal requirements for efficient use of personnel and equipment, record keeping, sample collection, storage, analysis, budget preparation, report writing, and planning for future work. Because the project involves many organizations external to the Laboratory, communication and coordination are especially important. This year we hosted a project leaders' meeting featuring technical presentations that emphasized explosion phenomenology in underground testing. We served as the lead organization in defining the source-term research goals for the Hydrology/Radionuclide Migration Project and contributed a number of position papers on other aspects of the project. We wrote the drilling plan for our next drilling project and initiated an effort to coordinate organizations involved in drilling, testing, and monitoring water wells or holes at the NTS.

Source-term research in FY1989 included sampling at the Cambric site and a new study of postshot debris. We have monitored the concentrations of tritium and fission products in the vicinity of the cavity at Cambric since 1975. The concentrations of tritium, ^{85}Kr , ^{90}Sr , and ^{137}Cs have continued to decrease at RNM-1 as water is pumped from the satellite well RNM-2S. In recent years, the concentrations of ^{85}Kr and tritium have been decreasing at similar rates, but the ratios of ^{90}Sr /tritium and ^{137}Cs /tritium are increasing. This is probably the result of leaching and desorption of the cationic materials, which partially offsets their removal by water transport. The concentrations of tritium, ^{85}Kr , and ^{90}Sr at RNM-1 are now approaching our limits of detection. Our initiation of a postshot debris study was motivated by a desire to learn whether there is significant separation of volatile and refractory materials in the cavity and chimney regions of underground nuclear test sites. Further, we would like to know the differences in leachability for elements present in these two regions. Postshot debris is routinely returned to Los Alamos for diagnostic purposes, and we wanted to know if this material could also be useful in our studies. We studied the postshot debris from two recent nuclear tests conducted in different underground environments. Samples collected from different regions at both test sites showed a marked differentiation of volatile and refractory materials. We developed a protocol for leaching these samples with groundwater and were able to measure some differences in leach rates for several elements. We calibrated our counting systems to accommodate these samples and adapted several computer codes for use in data analysis.

Transport studies conducted at Los Alamos this past year included our continued monitoring of the effluent from the RNM-2S well at Cambric, transmission electron microscope analysis of colloids collected from the Cambric site, and the first measurement of ^{99}Tc in Cambric waters. The shape of the tritium elution curve will provide researchers with data for transport model validation. We have pumped ~88% of the source term tritium out of the RNM-2S well, but only ~40% of the source term ^{85}Kr . Though we have pumped more than $1.5 \times 10^7 \text{ m}^3$ of water from this well, we have seen no ^{90}Sr or ^{137}Cs at RNM-2S. The nature and origin of colloids in groundwater at the Nevada Test Site has long been a subject of interest to the Hydrology/Radionuclide Migration Project. We recently isolated colloidal material from the RNM-1 well by ultrafiltration and then analyzed the colloids by transmission electron microscopy. Part of the material was crystalline and appeared to be aluminum silicates derived from native rock; the other part was an amorphous material containing calcium and phosphorous. We believe this latter material is a residue from drilling fluids that were introduced during construction of the RNM-1 well. The material's elemental composition may enable us to distinguish between "natural" colloids and those introduced with

drilling fluids. Isotopic composition might provide another means of making this distinction. We explored the use of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios to characterize two kinds of drilling mud and one representative tuff. The samples analyzed had distinctive strontium isotopic ratios, but we found that these ratios can vary considerably between various tuffs and even within an individual tuff unit. Therefore, we concluded that this approach would not be very useful. In another effort to attack this problem we used a geochemistry speciation code to calculate the mineral compositions that could precipitate from RNM waters. The results of these calculations could be used to eliminate certain possible mineral compositions; however, because a large number of other possibilities remain, this approach may have limited applicability for our work.

Pertechnetate ion is generally thought to be a conservative tracer for groundwater movement, but it is difficult to measure at very low concentrations. At Los Alamos, negative ion mass spectrometry is being used to measure ^{99}Tc . We recently obtained the first measurement of the ^{99}Tc concentration in a RNM-1 water sample collected in 1975. This technique appears to be sufficiently sensitive that we could analyze RNM-2S water samples.

Los Alamos personnel cooperated in hydrologic testing of the hole UE4t during this past year. Straddle packer tests confirmed the presence of a pressurized zone below the water table, and the pressure was great enough to cause hydraulic fracturing during the tests. The origin and characteristics of this pressurized aquifer are of considerable interest to the weapons testing community at the NTS.

During FY1989, Los Alamos was the lead organization for planning Hydrology/Radionuclide Migration Project research activities in the Yucca Flat area of the NTS. As part of our planning effort, we wrote the first draft of a drilling plan for the next hole in this area (UE3e#4). This hole will cost ~\$1.6M to drill and should yield a significant amount of data related to radionuclide transport and local hydrology.

I. INTRODUCTION

This report describes the work accomplished by Los Alamos National Laboratory personnel during FY1989 as participants in the Hydrology/Radionuclide Migration Project (HRMP). This project, funded by the US Department of Energy (DOE), supports research on radionuclide movement underground at the Nevada Test Site. We seek to determine the extent to which such movement occurs and the mechanisms that are operative in the movement. Organizations that contribute to HRMP research include Los Alamos National Laboratory (Los Alamos), Lawrence Livermore National Laboratory (LLNL), the US Geological Survey (USGS), the Desert Research Institute (DRI), and various support organizations at the NTS. The Nevada Operations Office of the US Department of Energy (DOE/NV) coordinates activities within the HRMP; technical review is provided by a steering committee of experts from relevant scientific disciplines.

The multidisciplinary research of the HRMP is organized into the following work elements: source term, transport, and local and regional hydrology. Most of the work described in this report involves research on source terms and transport. Los Alamos personnel are measuring the types and amounts of radionuclides and other hazardous materials (heavy metals and organics) present underground after a nuclear test and determining whether these materials move away from the immediate location of the test site. The emphasis at LLNL is in these same areas, whereas our colleagues at USGS and DRI concentrate more on hydrologic work. Within Los Alamos there are several divisions that contribute to HRMP research, including the Isotope and Nuclear Chemistry Division, the Field Test Division, and the Earth and Environmental Sciences Division. There is an obvious commonality of interest between the HRMP and the Yucca Mountain Project, and many Los Alamos personnel conduct research pertinent to both.

This report documents work performed by Los Alamos personnel and contains data generated during FY1989; previous data are included only if they are needed to help interpret recent results. The data presented are generally our own, and the data resulting from interorganizational cooperation will be identified. We have published a number of journal articles¹⁻³ and laboratory reports⁴⁻¹⁶ on various aspects of our past work in this area.

II. PROGRAM MANAGEMENT

The HRMP involves many people from different geographically situated organizations, so it is particularly important to establish mechanisms for coordinating our activities within the program. These mechanisms include rotating project leaders' meetings between participant organization locations, establishing lead organizations for specific HRMP projects, and assigning responsibility to define and identify programmatic objectives. During the past fiscal year, Los Alamos hosted a project leaders' meeting at which our staff gave technical presentations concerning weapons testing, environmental effects of this testing, and underground explosion phenomenology. Los Alamos served as the lead organization for HRMP research in the Yucca Flats area of the NTS. Research activities included packer testing at UE4t and preparing the drilling plan for the exploratory hole UE3e#4.

At the request of DOE/NV management, we organized a Hole Utilization Working Group with representatives from organizations involved in the drilling, testing, and monitoring of holes and wells at the NTS. Los Alamos personnel were responsible to the steering committee for defining HRMP goals and objectives of the source term task. We also provided a complete set of Los Alamos documents (journal articles and reports from the HRMP) to the new DOE/NV Coordination and Information Center in Las Vegas. A duplicate set of documents was sent to the Pacific Northwest Laboratory task force, which was preparing an updated bibliography of NTS publications pertinent to the HRMP. The Appendix of this report contains lists of HRMP meetings attended by Los Alamos personnel and the documents we prepared during the past fiscal year.

III. SOURCE TERM

A. CAMBRIC RNM-1 (J. Drake, W. D. Efurd, S. Maestas, and J. L. Thompson)

In 1965 the nuclear test Cambric was conducted in the tuffaceous alluvium of Frenchman Flat at the NTS. This became the first field study site for the HRMP. Re-entry drilling in 1974 revealed that most of the radionuclides associated with the test were still in the cavity region. A satellite well, RNM-2S, was drilled 91 m away and water was pumped out at 2.3 m³/min to induce a hydraulic gradient at the cavity. Pumping has continued since 1974 with only short interruptions. We monitor the concentration of radionuclides in the water at the cavity (RNM-1) and in the pumped effluent from RNM-2S.

Water samples collected from the RNM-1 well on an annual basis consist of 208-l samples in polyethylene-lined barrels, 2-l samples in air-tight stainless steel tubes, and a number of additional samples of varying size collected in 20- to 0.5-l polyethylene bottles. The large-volume samples are evaporated and the residue is measured with Ge(Li) detectors to determine the presence of gamma-emitting radionuclides. The 2-l samples are analyzed on a vacuum line for their ⁸⁵Kr content. The variable-size samples are analyzed for tritium, ⁹⁰Sr, and ¹³⁷Cs. A more complete description of our sampling and counting protocols is given in Ref. 13.

In Table I we list the tritium content of samples collected on 8/29/89 during the annual pumping of the RNM-1 well. In Tables II and III we show how the concentrations of ⁸⁵Kr, ⁹⁰Sr, and ¹³⁷Cs have changed relative to tritium since 1975. All of the radionuclides we have monitored at RNM-1 have decreased dramatically in concentration as water has been pumped out from RNM-2S. The decrease in concentration of ⁸⁵Kr relative to tritium has become fairly constant in recent years, although the atom ratio of ⁸⁵Kr/tritium remains somewhat higher than the source term value. Apparently krypton and tritium concentrations are decreasing at the cavity at similar rates; however, the arrival of tritium at RNM-2S slightly precedes that of ⁸⁵Kr (see Figs. 1 and 2), and the atom ratio ⁸⁵Kr/tritium at RNM-2S is less than the source term value (see Sec. IV.A). The apparent loss of krypton relative to tritium may be a result of sorption (perhaps on zeolites in the alluvial material) or another loss mechanism such as escape to the atmosphere. In contrast with the situation for krypton, the tritium concentration is decreasing in the cavity more rapidly than the concentrations of strontium and cesium, so the atom ratios for ⁹⁰Sr/tritium and ¹³⁷Cs/tritium are

Table I. Tritium Concentration in RNM-1 Water (1989)

Identification Number	Volume Pumped (m ³)	Tritium Concentration (10 ⁻⁶ mCi/ml)	
		On Collection Date 8/29/89	Corrected to Cambric T ₀ 5/14/65
V-1	10	0.20	0.77
V-2	21	0.24	0.93
V-3	31	0.21	0.84
V-4	43	0.19	0.75
V-5	53	0.25	1.0
V-6	63	0.28	1.1
V-7	80	0.26	1.0

Table II. Tritium and ^{85}Kr Concentrations in RNM-1 Water (1989)

Zone	Date	Volume Pumped ^a [x 10 ⁶ (m ³)]	Concentration ^b	Concentration	^{85}Kr /Tritium	
			Tritium ($\mu\text{Ci}/\text{mL}$)	^{85}Kr ($\mu\text{Ci}/\text{mL}$)	Atom ratio ^c (x 10 ⁴)	Ratio ^d
Original-Zone IV	08/08/75	0.	1.5×10^{-1}	3.1×10^{-5}	1.8	1.5
Original-Zone V	08/14/75	0.	3.8×10^{-2}	6.1×10^{-6}	1.4	1.2
Zone IV + V	10/04/77	1.17	3.2×10^{-3}	3.4×10^{-5}	92	75
Zone IV + V	11/30/77	1.34	2.0×10^{-3}	2.7×10^{-6}	12	10
Zone IV + V	09/04/79	3.50	2.6×10^{-4}	2.5×10^{-7}	8.5	7
Zone IV + V	10/05/81	5.89	1.4×10^{-4}	8.3×10^{-8}	5.0	4
Zone IV + V	04/01/82	6.48	7.7×10^{-5}	3.0×10^{-8}	3.3	3
Zone IV + V	04/06/83	7.67	3.3×10^{-5}	1.3×10^{-8}	3.4	3
Zone IV + V	04/24/84	8.91	2.2×10^{-5}	1.0×10^{-8}	4.1	3
Zone IV + V	04/10/85	10.0	1.3×10^{-5}	1.2×10^{-8}	7.8	6
Zone IV + V	05/07/86	11.1	5.8×10^{-6}	2.1×10^{-9}	3.2	3
Zone IV + V	03/25/87	12.2	3.6×10^{-6}	1.7×10^{-9}	4.0	3
Zone IV + V	05/10/88	13.5	2.2×10^{-6}	—	—	—
Zone IV + V	08/29/89	15.1	9×10^{-7}	(analysis in progress)		

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

^bValues corrected to Cambrian zero time.

^c 1.81×10^{13} atoms ^{85}Kr = 1 μCi ; 2.08×10^{13} atoms tritium = 1 μCi .

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

increasing. Desorption and dissolution of strontium and cesium in the cavity region will tend to counter the removal of these materials as dissolved species; presumably, at some point a steady state will be reached in which the concentrations will not change at all with time.

Concentrations are now so low that measurement of tritium, ^{85}Kr , and ^{90}Sr is difficult; only ^{137}Cs is still present at concentrations well above our detection limits. As discussed in Ref. 13, our counting procedures enable us to measure tritium at concentrations above ~1 pCi/mL, and ^{85}Kr , ^{90}Sr , and ^{137}Cs (by a radiochemical method) above $\sim 10^{-2}$ pCi/mL. We can detect ^{137}Cs down to $\sim 10^{-5}$ pCi/mL by using our boildown procedure. The most recent data in Tables I, II, and III suggest that by next year only the concentration of ^{137}Cs can be monitored at RNM-1 with good precision.

Zone	Date Collected	Volume Pumped ^a (10^6 m^3)	Concentration ^b			Atom Ratio ^c ($\times 10^4$)	
			Tritium ($\mu\text{Ci}/\text{mL}$)	^{90}Sr ($\mu\text{Ci}/\text{mL}$)	^{137}Cs ($\mu\text{Ci}/\text{mL}$)	$^{90}\text{Sr}/\text{T}$	$^{137}\text{Cs}/\text{T}$
Original-Zone IV	08/08/75	0.	1.5×10^{-1}	2.7×10^{-6}	4.5×10^{-7}	0.4	0.07
Original-Zone V	08/14/75	0.	3.8×10^{-2}	1.1×10^{-7}	9.0×10^{-8}	0.07	0.06
Zone IV + V	10/04/77	1.17	3.2×10^{-3}	4.6×10^{-7}	1.1×10^{-7}	3	0.8
Zone IV + V	11/30/77	1.34	2.0×10^{-3}	2.7×10^{-7}	2.2×10^{-7}	3	3
Zone IV + V	09/04/79	3.50	2.6×10^{-4}	9.1×10^{-8}	4.5×10^{-8}	8	4
Zone IV + V	10/05/81	5.89	1.4×10^{-4}	3.1×10^{-8}	2.6×10^{-8}	5	5
Zone IV + V	04/01/82	6.48	7.7×10^{-5}	2.7×10^{-8}	3.0×10^{-8}	8	9
Zone IV + V	04/06/83	7.67	3.3×10^{-5}	—	2.6×10^{-8}	—	19
Zone IV + V	04/24/84	8.91	2.2×10^{-5}	1.7×10^{-8}	2.0×10^{-8}	18	22
Zone IV + V	04/10/85	10.0	1.3×10^{-5}	1.1×10^{-8}	2.0×10^{-8}	20	38
Zone IV + V	05/07/86	11.1	5.8×10^{-6}	1.8×10^{-8}	2.0×10^{-8}	72	100
Zone IV + V	03/25/87	12.2	3.6×10^{-6}	3×10^{-9}	1.9×10^{-8}	20	130
Zone IV + V	05/10/88	13.5	2.2×10^{-6}	4×10^{-9}	1.7×10^{-8}	40	190
Zone IV + V	08/29/89	15.1	9×10^{-7}	2×10^{-8}	1.2×10^{-8}	500	300

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

^bValues corrected to Cambrian zero time.

^c 4.81×10^{13} atoms $^{90}\text{Sr} = 1 \mu\text{Ci}$; 5.09×10^{13} atoms $^{137}\text{Cs} = 1 \mu\text{Ci}$.

Fig. 1. Tritium concentration in water pumped from RNM-2S vs volume pumped.

Fig. 2. The ^{85}Kr concentration in water pumped from RNM-2S.

B. ANALYSIS OF TEST DEBRIS (J. L. Thompson)

As a result of volatilization and condensation processes taking place during the cavity formation in an underground nuclear test, there may be some separation of volatile and refractory elements.¹⁷ We are attempting to obtain more field data on the spatial distribution of fission products in nuclear explosion cavities. We also want to discover what differences exist in the solubilities of volatile and refractory elements because dissolution may be the time-controlling step in the transport of fission products away from the cavity region. This year we began exploring ways to acquire such data by using material available from other Los Alamos activities at the NTS. Postshot debris samples are routinely returned to Los Alamos for radiochemical analyses; these analyses give information about the performance of the test device. Typically, not all the debris returned is required for diagnostic purposes, and this excess material is available for other use. Our initial work was to test the feasibility of using excess postshot debris and to develop some protocols for future experiments. Our counters had not been calibrated for the sample configurations we used, so the data are reported only on a relative basis. Debris from two recent tests (Shot A and Shot B) were studied.

1. Shot A

This test device was exploded at a depth of 350 m in zeolitized, ash-flow tuff above the water table. The tuff had a 13% (by weight) water content and 40% porosity; it was 55% saturated. Sample A1 was representative of the puddle material. We detected ⁹⁵Zr, ⁹⁵Nb, ¹⁰³Ru, ¹²⁴Sb, ¹³¹I, ¹⁴⁰Ba, ¹⁴¹Ce, ¹⁴⁴Ce, and ¹⁴⁷Nd in this sample. This result was in agreement with the diagnostic analysis; however, the more sensitive counting techniques employed in the latter case also detected several other radionuclides at low concentrations. Sample A2 was collected ~9 m below sample A1 from a fissure thought to be enriched with volatile material. We detected the same suite of radionuclides as in Sample A1, plus ¹³²Te and ¹³⁶Cs. The comparison of these radionuclides is shown in Table IV.

2. Shot B

This shot was fired at a depth of 622 m in unsaturated lava with 4.5% (by weight) water content, 11.5% porosity, and 88% saturation. Sample B1 was obtained from the puddle region. The radionuclides identified were ⁹⁵Zr, ⁹⁵Nb, ⁹⁹Mo, ¹⁰³Ru, ¹²⁴Sb, ¹²⁷Sb, ¹³²Te, ¹³¹I, ¹⁴⁰Ba, ¹⁴¹Ce, ²³⁷U, and ²³⁹Np. Sample B2 was taken from ~21 m above the puddle. In sample B2 we did not detect the refractory elements ⁹⁵Zr, ⁹⁵Nb, and ¹⁴⁷Nd, but these were found in low concentrations by the diagnostic analyses. The results of our comparisons are shown in Table V.

Table IV. Relative Amounts of Radionuclides in Samples from Shot A

Radionuclide	A1/A2
⁹⁵ Zr, ⁹⁵ Nb	36
¹⁴⁷ Nd	33
¹⁴⁰ Ba	7.4
¹⁰³ Ru	1.3
¹⁴¹ Ce	1.2
¹³¹ I	1.0

Table V. Relative Amounts of Radionuclides in Samples from Shot B

Radionuclide	B1/B2
⁹⁵ Zr, ⁹⁵ Nb, ¹⁴⁷ Nd	very high
²³⁹ Np	240
⁹⁹ Mo	210
¹⁴¹ Ce	130
¹⁴⁰ Ba	45
¹⁰³ Ru	44
¹³² Te	15
¹³¹ I	5.6
¹²⁴ Sb, ¹²⁷ Sb	1-10

Comparison of the data in Tables IV and V shows that the samples from Shot B had more differentiation between refractory and volatile elements than did the samples from Shot A. More importantly for our work, however, both sets of samples proved satisfactory for measuring the existence of this differentiation. We are encouraged to continue to use excess postshot debris to quantify the spatial separation of volatile and refractory elements in the cavity/chimney region. The second aspect of these studies is the determination of relative rates of dissolution in groundwater for the elements present after the device has detonated. For our preliminary studies with postshot debris, we devised leaching experiments using a water-to-rock ratio equal to that used for most of the Los Alamos radionuclide sorption studies.¹⁸ We also examined the effect of increasing the temperature. We used the protocols listed below for these experiments.

- (1) A weighed sample of finely ground shot debris (~1 g) is placed in a screw-cap centrifuge tube with ~20 ml of water from the J-13 well at the NTS.
- (2) The rock and water mixture is shaken in a constant temperature bath for a period of a few weeks or a few months.
- (3) The mixture is centrifuged to separate the phases and an aliquot of the water is filtered through a 50-nm Nuclepore filter to remove any fine particulates.
- (4) A 5-ml fraction of the separated water is counted on a calibrated high-purity germanium counter, and the gamma spectrum is analyzed using the computer code RAYGUN.
- (5) The separated water is reintroduced to the centrifuge tube and shaking is resumed for the next period. The results of these leaching experiments on debris from Shot A and Shot B are shown in Table VI. The time lapse between the shot and the counting is designated as " Δt ."

These preliminary leaching experiments indicate that differences in sample dissolution may be observed for elements distributed in different zones of the cavity/chimney region. The concentrations of dissolved materials were quite low, and more exact computer codes (for example, GAMANAL and CLSQ) should be used to better quantify the dissolved radionuclides. The number of gamma-emitting nuclides detectable after several months appears to be rather limited. However, increasing the temperature during leaching does appreciably increase the concentration of the dissolved species.

Table VI. Leaching Experiments on Debris from Shot A and Shot B

Sample	Weight Sample (g)	Volume Water (ml)	Shaking Time (days)	Temperature (°C)	Δt (days)	Radionuclides Identified	Relative Concentration (%)
A1	1.421	27.5	20	20	53	^{124}Sb , ^{131}I	100
			145	20	199	^{124}Sb	34
			7	70	211	^{124}Sb	44
A2	1.276	28.4	20	20	53	^{103}Ru , ^{124}Sb , ^{131}I	100
			145	20	199	^{124}Sb	37
			7	70	211	^{103}Ru ^{124}Sb	20 34
B1	1.262	25.2	14	20	33	^{95}Nd , ^{99}Mo , ^{237}U , ^{103}Ru , ^{124}Sb , ^{131}I	100
			117	20	154	^{95}Nd	47
			7	70	165	^{95}Nd	68
B2	1.040	20.8	14	20	33	^{103}Ru , ^{124}Sb , ^{131}I , ^{132}Te , ^{140}Ba	100
			117	20	154	^{103}Ru	20
						^{124}Sb	35

It became apparent during these preliminary studies that it would be useful to calibrate the counters so that solid and liquid samples could be directly compared and the fraction leached could be computed for each radionuclide. The calibration of our counters for these samples was done using a mixed gamma standard purchased from Amersham. This standard is certified by the British Calibration Service and is traceable to the US National Bureau of Standards (now the National Institute of Standards and Technology). The counters used in this work are high-purity germanium detectors coupled with multichannel analyzers and automated sample changers. The spectra are analyzed using the computer codes GAMANAL and CLSQ. The solid samples are contained in a plastic vial ~9 mm in diameter and 9 mm high, that holds 0.2 to 0.3 g of the ground rock. These vials are placed in plastic screw-cap containers that are held in a lucite plug; this plug fits on the sample changer and can be positioned at standard counting geometries. The calibration of this counting system was carried out by adding known amounts of the standard to nonradioactive dirt. A counter efficiency curve for gamma rays in the energy range of 60 to 1836 KeV was constructed for each of several sample-counter geometries. Following the calibration, we are able to measure the number of atoms/g in our solid samples (and the number of atoms/ml in liquid samples) for each of the detectable radionuclides. Thus, we can do absolute measurements of the gamma-emitting radionuclides in the debris samples and in the leachate. This should improve our ability to both analyze the components in postshot debris and predict the degree to which these components can be leached by groundwater.

IV. TRANSPORT

A. CAMBRIC RNM-2S (S. Maestas and J. L. Thompson)

The Cambria field test is described in Sec. III.A. We collect water samples from the pumped well RNM-2S on a monthly basis. The large-volume (208-l) samples are boiled down for gamma counting or stored for future analyses. The small-volume samples are analyzed for tritium, and the 2-l samples in stainless steel tubes are analyzed for ^{85}Kr . The results of our tritium analyses during the past fiscal year are presented in Table VII; the tritium and krypton analyses are in Table VIII. Cumulative data for the elution of tritium and krypton are shown in Figs. 1 and 2, respectively. As we mentioned earlier in this report, the krypton is apparently slightly delayed relative to tritium in traveling from the cavity to the pumped RNM-2S well. Also, the ^{85}Kr /tritium ratio is consistently lower at the RNM-2S well than at the RNM-1 well. At the time of the last sampling, we had pumped out ~88% of the Cambria source term tritium and ~40% of the source term ^{85}Kr . It will be very interesting to see if this "missing" krypton appears with continued pumping of RNM-2S.

B. COLLOID STUDIES (S. Goldstein and J. Fabryka-Martin)

We are concerned about the origin of colloids in water samples collected at the NTS and would like to distinguish between colloids originating in the native rock and those introduced with drilling fluids. If the strontium isotopic composition of the drilling fluids and the rock were sufficiently different, this might serve as the basis for determining the origin of a collected colloid sample. To test this possibility, we had two kinds of drilling mud and a representative tuff (YM22-Topopah Springs) analyzed for strontium, rubidium, and the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio. The analysis was done by the Chempet Research Corporation, Moorpark, California. The results are shown in Table IX.

These data seem to suggest that bentonite could readily be distinguished from tuff. However, further investigation revealed that this might not be true in many situations. One recent study¹⁹ found that the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio varied from 0.7160 in the Topopah Spring tuff to 0.7089 in an older tuff unit. In another study,²⁰ systematic variations of the strontium isotope ratios within individual

Table VII. Tritium Analyses of RNM-2S Water Samples

Identification Number	Date Collected	Volume Pumped (10^6 m^3)	Tritium Concentration ^a ($10^{-4} \mu\text{Ci/ml}$)	
			At Counting Time	At T_0 5/14/65
G 1485	10/11/88	14.024	5.39	20.4
G 1486	11/15/88	14.134	5.23	19.8
G 1488	01/19/89	14.348	5.12	19.4
G 1489	01/26/89	14.374	4.99	19.2
G 1490	02/23/89	14.464	4.94	19.0
G 1491	03/20/89	14.546	4.89	18.8
G 1492	04/24/89	14.657	4.76	18.5
G 1493	05/16/89	14.728	4.76	18.5
G 1494	07/14/89	14.919	4.58	17.8
G 1495	08/16/89	15.026	4.44	17.4
G 1496	09/15/89	15.124	4.34	17.0

^aStandard deviation of counting data <1%.

Table VIII. Tritium and ^{85}Kr Analyses of Pressurized Water Samples from RNM-2S ^a

Identification Number	Date Collected	Pumped (10^6 m^3)	Tritium Concentration ($10^{-3} \mu\text{Ci/mL}$)	^{85}Kr Concentration ($10^{-7} \mu\text{Ci/mL}$)	$^{85}\text{Kr}/\text{Tritium}$	
					Atom Ratio ^b ($\times 10^5$)	Ratio ^c
434-1-88-016	08/02/88	13.797	2.09	1.49	6.2	0.51
434-1-88-017	09/01/88	13.895	2.04	1.48	6.3	0.52
434-1-88-019	10/05/88	14.005	2.00	1.48	6.4	0.52
434-1-88-021	11/04/88	14.099	2.02	1.26	5.4	0.44
434-1-88-024	12/02/88	14.190	1.98	1.52	6.7	0.55
434-1-88-001	01/26/89	14.371	1.83	1.27	6.0	0.49
434-1-88-003	02/02/89	14.394	1.82	1.48	7.1	0.58
434-1-88-006	03/14/89	14.523	1.80	1.16	5.6	0.46
434-1-88-007	04/06/89	14.598	1.77	1.30	6.4	0.52
434-1-88-010	05/02/89	14.684	1.80	1.30	6.3	0.52
434-1-88-011	06/01/89	14.780	1.79	0.98	4.8	0.39
434-1-88-013	07/06/89	14.892	1.77	1.44	7.1	0.58

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

^b 1.81×10^{13} atoms $^{85}\text{Kr} = 1 \mu\text{Ci}$; 2.08×10^{13} atoms tritium = $1 \mu\text{Ci}$.

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

Table IX. Analysis of Drilling Fluids and Tuff

Sample	$^{87}\text{Sr}/^{86}\text{Sr}$	Sr(ppm)	Rb(ppm)
YM22	0.71673	29.5	146.4
IMV #6 Sepiolite	0.71219	352	47.7
API #1-2 Bentonite	0.70946	164.5	40.6

ash-flow units were observed. If each stratigraphic unit has its own ratio and this ratio can change within the unit, then identifying the origin of a particular sample could be very difficult. The problem may be particularly difficult in the alluvium in Frenchman Flat, which contains detrital limestone, dolomite, quartzite, rhyolite, tuff, and argillite.²¹ This mix of strontium sources will probably vary spatially in plan view and with depth, depending on local drainage patterns. Yet another complication is the possibility that colloids may exchange bound strontium with dissolved strontium in the groundwater—thus losing their original isotopic tag. We have concluded that only in special circumstances will the strontium isotopic ratio be useful in distinguishing natural colloids from colloids originating in drilling fluids.

The RNM-1 well at the Cambrian site is annually pumped to monitor the concentrations of radionuclides in the region of the cavity (refer to Sec. III.A). We normally pump out about 80 m³ of water during the sampling. We collected ~80 l of water for colloid analysis toward the end of the 1989 sampling exercise at RNM-1 and stored it in a polyethylene container. After returning to the laboratory, we filtered the sample in a Millipore Pellicon ultrafiltration system with a 100 000-MW filter (~6.9-nm pore diameter). A colloid concentrate of 250 ml was saved, from which samples were prepared for transmission electron microscopic analysis. Portions of the concentrate were centrifuged onto copper grids covered with a carbon film. A blank was similarly prepared by using a "concentrate" from distilled water processed through the filtration system. The results of the analysis are shown in Table X. The distilled water blank indicated that the carbon film was quite clean; only minor amounts of silicon and iron were detectable. The colloidal material was amorphous or crystalline. Crystalline particulates contained silicon, aluminum, potassium, and magnesium as the major elements; these probably are clays derived from the native rock. The amorphous particulates often contained calcium and phosphorous as major components. We hypothesize that this material is similar to apatite in composition; for example, $\text{Ca}_5(\text{F}, \text{Cl}, \text{OH})(\text{PO}_4)_3$. It is probable that this material was introduced as drilling fluid during construction of the well, because the natural phosphate content of NTS waters is quite low.¹⁸

Another way to screen the possible colloids in groundwaters at the NTS is by calculating the mineral saturation indices. This approach establishes the presence (or absence) of a thermodynamic basis for colloids of a specific mineralogy. It is limited by the quality of the available information about water composition at the particular site. To apply this approach at the Cambrian site, we used the geochemistry speciation code PHREEQE (Ref. 22) to calculate the saturation indices of various minerals with elemental compositions similar to those in Table X. These calculations were based on water analyses²¹ from well 5B, which is in the vicinity of the Cambrian site. The results of these calculations are shown in Table XI. Saturation indices greater than zero indicate precipitation (hence, possible colloid formation); those less than zero indicate mineral dissolution. These

Table X. Colloid Analysis by Transmission Electron Microscopy

File Number	Sample	Elements		Structure	Size (μm)
		Major	Minor		
70	area scan	Si	Ca,S,P,K,Fe		
71	particle	Ca,P	Mg	amorphous	0.05-0.1
72	around #71	Si,Ca,P,S	Fe,Mg	amorphous	
73	particle	Mn,Si	Ca,S	amorphous	0.05-0.1
74	particle	Ca,P	Mg	amorphous	0.3-0.4
75	particle	Si,K,Al	Na	crystalline	0.5
76	around #75	Si,Al,Mg	Fe,K	amorphous	
77	particle	Si,Al	Ca,Fe,K	crystalline	0.5
78	particle	Si,Al,K	Ca,Fe	crystalline	
79	particle	Ca,P	Mg	amorphous	0.3
80	particle	Ca,As	Si,Mg	amorphous	0.1

Table XI. The PHREEQE Calculations of Saturation Indexes

Mineral	Saturation Index	Formula
Carbonate apatite	12.8	$\text{Ca}_{9.5}\text{Mg}_{0.14}\text{F}_{2.48}(\text{PO}_4)_{4.8}(\text{CO}_3)_{1.2}$
Hydroxylapatite	-1.5	$\text{Ca}_5(\text{PO}_4)_3\text{OH}$
Fluorapatite	7.6	$\text{Ca}_5\text{F}(\text{PO}_4)_3$
Microcline (K-spar)	4.4	KAlSi_3O_8
Sanidine	3.9	KAlSi_3O_8
Anorthite	-0.2	$\text{CaAl}_2\text{Si}_2\text{O}_8$
Muscovite (K-mica)	10.2	$\text{KAl}_2(\text{AlSi}_3\text{O}_{10})(\text{OH})_2$
Pyrophyllite	7.9	$\text{Al}_2\text{Si}_4\text{O}_{10}(\text{OH})_2$
Laumontite	4.8	$(\text{Ca},\text{Na})_7\text{Al}_{12}(\text{Al},\text{Si})_2\text{Si}_{26}\text{O}_{80} \cdot 25 \text{H}_2\text{O}$
Kaolinite	6.5	$\text{Al}_4(\text{Si}_4\text{O}_{10})(\text{OH})_8$
Nontronites	21-28	$\text{Fe}_4\text{AlSi}_8\text{O}_{20}(\text{OH})_4$
Illite	4.4	mica-like clay mineral
Montmorillonite	3.9	$(\text{Al},\text{Mg})_8(\text{Si}_4\text{O}_{10})_3(\text{OH})_{10} \cdot 12 \text{H}_2\text{O}$
Chlorites	5-31	$(\text{Mg},\text{Fe})_3\text{Si}_4\text{O}_{10}(\text{OH})_2\text{Mg}_3(\text{OH})_6$

calculations indicate conditions favorable for precipitation of fluorapatite, carbonate apatite, certain feldspars, micas, zeolites, and clays. It is apparent that a wide range of mineralogic compositions are thermodynamically possible and could be sources for colloidal material. Indeed, the calculational approach may be of limited utility because it does not sufficiently limit the number of possible materials for colloid formation.

C. TECHNETIUM-99 ANALYSIS (A. E. Ogard and D. J. Rokop)

The Los Alamos HRM was not funded to do technetium analyses in FY1989; however, when another project offered to run samples for us, we were happy to provide water from the Cambric and Cheshire cavities. Sample preparation involves separation of pertechnetate on an anion exchange column, elution with nitric acid, drying, sublimation from aqua regia, and drying in the presence of HI. A detailed description of this technique has been published recently.²⁴ Analysis is done by negative ion mass spectroscopy. Table XII shows the results of the Los Alamos analyses, as well as a comparison with results by beta counting from LLNL (Ref. 23).

We used 1 l of Cambric cavity water in this analysis, but we estimate that the sensitivity is such that only 1 ml was required. This methodology will allow us to analyze ⁹⁹Tc in water from the RNM-2S well or from other sources where the technetium is below the limits of detection by beta counting.

Table XII. Concentrations from ⁹⁹ Tc Analyses		
Sample	Concentration (pg/ml)	
	LANL	LLNL
Cheshire (B-57)	3.97	2.95
Cambric (D-13b)	0.0134	<0.05 (upper limit only)

V. LOCAL HYDROLOGY (W. L. Hawkins, T. M. Mahevc, and D. A. Trudeau)

One task of the HRMP research program at the NTS is to examine the effects of underground explosions on the local hydrology. During FY1989, Los Alamos participated in straddle packer testing in the hole UE4t, which had been drilled adjacent to a recent test. This work, a cooperative effort between USGS, DRI, and Los Alamos, was described in detail in a recent paper²⁵ and is only summarized here. Hole UE4t was drilled in early 1987 to investigate hydrologic and radionuclide migration following the GASCON underground test conducted in hole U4t, 170 m to the north. There have been 19 underground tests conducted within 1500 m of UE4t in the same structural block. The 0.25-m-diam hole was rotary drilled using air foam to a total depth of 736 m. Hole UE4t is in a north-trending geologic structural block that is bounded on the east by the Yucca fault and on the west by the Topgallant fault. The strata dip is 17° to the west.

Lithologic units penetrated by UE4t include 131 m of alluvium overlying ash-flow, air-fall, and bedded tuff units and minor tuffaceous sandstones. Below 308 m, the tuffs are moderately to heavily zeolitized and are considered part of the tuff-confining hydrologic unit. Data gathered during drilling indicate the presence of elevated fluid pressures below the water table. Before the packer testing, the fluid level had risen in this uncased hole to a depth of only 268 m. Estimated depth to the water table is 506 m. Straddle packer testing was conducted in UE4t during January and

February of 1989. The packer testing assembly was designed to make pressure measurements above, below, and between the packers. Testing confirmed the presence of elevated fluid pressures in formations below the estimated water table. Hydraulic fracturing was observed during some of the tests. A test of the portion of the hole below 629 m resulted in fluid rising to within 106 m of the surface, indicating a formation pressure of ~755 psia.

VI. DRILLING UE3e#4 (A. E. Ogard and J. L. Thompson)

For several years the HRMP has been studying a site in Area 3 at the NTS at which radionuclide migration has been detected.^{10,11} We have identified a variety of radionuclides that have apparently moved; these radionuclides seem to be confined within a narrow region, which suggests that movement occurred within a fracture. Several attempts to core through this fracture have proved futile. Core recovered during drilling of UE3e#2 contained no radioactivity (although tritium was present in the water at depth in the hole). Hole UE3e#3 caved in before coring could be accomplished. We had hoped to drill another hole, UE3e#4, in this area during FY1989, but budget constraints postponed the actual drilling. We did prepare a drilling plan for this hole; the plan calls for drilling a vertical hole 17-1/2-in. diam to 1650-ft depth, and 9-7/8-in. diam to 1950 ft. The drilling is accomplished using air foam, and the casing is set in the upper section of the hole. Coring is done in the interval from 1950 to 2250 ft, then the hole is overdrilled to 2300 ft. Full suites of geophysical logs are run in both the upper and lower sections of the the hole. A pump test and straddle packer tests are run in the hole. It is completed with a lockable wellhead cover so it may be accessed for future sampling. The cost estimate (including a 20% contingency fund) for this drilling is ~\$1.6M. It is understood that alterations in the plan will occur during the course of actual drilling.

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APPENDIX

ADDITIONAL ACTIVITIES AND PROJECT CONTRIBUTIONS

HRMP Meetings Attended During FY1989

LLNL, Livermore, California	October 27-28, 1988
LANL, Los Alamos, New Mexico	February 16-17, 1989
DRI, Reno, Nevada	May 4-5, 1989
USGS, Las Vegas, Nevada	August 28-29, 1989
Additional Meeting: Hole Utilization Working Group, Mercury, Nevada	December 15, 1988

HRMP Documents Prepared During FY1989

"Laboratory and Field Studies Related to the Hydrology/Radionuclide Migration Project, October 1, 1987—September 30, 1988," Los Alamos National Laboratory report LA-11595-PR (July 1989).

HRMP Summary Sheets, FY1989

Drilling Plan UE3e#4, November 1988

HRMP Budget Projections, FY1990

Critique of Eddy *et al.* update of the Borg report, January 1989

Critique of LLNL "Draft Proposal for Cheshire Study Plan," January 1989

"Migration of Fission Products at the Nevada Test Site: Detection with an Isotopic Tracer," abstract submitted to Migration '89, Monterey, California, November 6-9, 1989

"Radionuclide Migration Studies at the Nevada Test Site," abstract submitted to 1989 International Chemical Congress of Pacific Basin Societies, Honolulu, Hawaii, December 17-22, 1989

Long-Range Plan for the HRMP, July 1989

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