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

October 1, 1989--September 30, 1990

Compiled and Edited by Joseph L. Thompson

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ABSTRACT

This annual report describes research conducted in FY 1990 by Los Alamos National Laboratory for the Hydrology/Radionuclide Migration Project. This multi-agency project measures the underground movement of radionuclides related to nuclear testing at the Nevada Test Site. This project continues the long-term experiment at the site of the Cambric nuclear test. Water pumped from a well adjacent to the explosion cavity continues to show decreasing amounts of tritium and ^{85}Kr , but no ^{137}Cs . Analyses of drillback debris shows a distinction between refractory and volatile materials in respect to both their location in the test cavity and their leachability with groundwater. We surveyed materials used during nuclear testing to evaluate any post-test hazard; we concluded that most such materials pose a minimal hazard. The Los Alamos drilling program provided an opportunity for us to sample a collapsed zone above the cavity of a test, which was fired 2 years ago. We continue our research in colloid characterization and in detection of low levels of ^{99}Tc in Nevada Test Site water. During FY 1990, we drilled a new hole in the Yucca Flat area to study radionuclide migration. This report also describes Los Alamos management and planning activities in support of this project.

EXECUTIVE SUMMARY

This report describes the FY 1990 research conducted by Los Alamos National Laboratory personnel for the Hydrology/Radionuclide Migration Project. Our project studies the environmental consequences of nuclear testing at the Nevada Test Site (NTS), especially the underground movement of radionuclides and other hazardous materials that reside underground after nuclear tests (source terms) and the movement of these materials through the geologic media surrounding them (transport). This annual report is the most recent in a series of reports and articles published over the 17 years of this project.

Program management at Los Alamos includes normal operational concerns, with special emphasis on cooperation between organizations both within and outside the Laboratory. During FY 1990, Los Alamos served as the coordinator for the HRMP drilling of the hole UE3e#4 at our study location in Area 3 of the NTS. We were the lead organization for a project that involved drilling into the chimney of an event located at U4u. In addition to this annual report, we have publicized our research through several international presentations, in which we discussed HRMP's unique opportunities for research on radionuclide behavior in the underground environment.

Our FY 1990 source-term research included analyses of Cambric RNM-1 water samples, analyses of postshot debris, evaluation of the materials used in underground nuclear testing, and re-entry drilling into the chimney at a test site. We wish to gain a better understanding of what materials—both radioactive and hazardous—are typically present after a nuclear test. We also seek to discover if the chemical state of these materials allows them to move with groundwater toward the accessible environment.

At the Cambric site, we have continued to sample water from the cavity region (RNM-1). During FY 1990, the activity levels of tritium, ^{85}Kr , and ^{90}Sr have diminished to background values; only ^{137}Cs could be measured with precision. After about 15 years of pumping water from the adjacent well (RNM-2S), we have greatly lowered the concentrations of the radionuclides in the cavity water. We can now detect only the nuclide for which our counters have the greatest sensitivity. At RNM-1, different transport behaviors have been caused by the different chemical properties of the four radionuclides; at present, we can fully account for only one of these radionuclides. This topic is mentioned again below as part of the discussion of transport.

In FY 1989, we began to analyze postshot debris shipped to Los Alamos for diagnostic purposes. This year we studied the debris from three shots; we looked for differences in the distribution of volatile and refractory materials as a function of the debris's location in the cavity/chimney complex, and noted differences in the leachability of radionuclides from the various locations. From the current data, it

appears that all elements—refractory and volatile—are more leachable when obtained at a distance from the puddle at the bottom of the cavity. This leachability pattern may reflect the materials condensing as light films at the top of the cavity or as thick layers at the bottom of the cavity. More data are needed to see if these initial trends continue. We would like to establish whether or not the location of an element within the cavity/chimney complex affects its contribution to the hydrologic source term, that is, to the source term of transportable materials.

Materials placed underground in the course of conducting a nuclear test often reside there after the test. We have sought to evaluate the hazards associated with materials that are not radioactive, but are present as a result of nuclear testing. Over 600 underground tests have been conducted at the NTS; about 28% of these were at or below the water table. We reviewed the underground nuclear test process, particularly emphasizing investigations of stemming and drilling materials, and the rack and device components. We identified the sources and records needed to garner this information, and tested our ability to assemble this information by inventorying the material used in the Amarillo event of 1989. We concluded that organic materials do not constitute a hazard, that most metals were present in such small concentrations as to pose no hazard, and that metals present in larger amounts, such as lead, would have to be in a soluble form before they could present a hazard.

Los Alamos drilling (J Division) presented us with a rare opportunity to drill into the collapsed zone above the cavity bottom without going all the way into the puddle region. This drilling enabled us to obtain material without possible contamination of samples by the intensely radioactive puddle material. J Division conducted this drilling program as a full-scale test of a new drillback rig. J-Division's willingness to drill at an HRMP site, thus facilitating our research, reflects the valuable cooperation that is very beneficial to our program. We logged the gamma radiation in the hole with a tool more sensitive than the type usually employed in drillbacks; with the gamma log guided our sampling, we obtained samples containing ^{137}Cs , ^{125}Sb , and ^{106}Ru . A tube was set in the hole to allow us to monitor the water level and to obtain water samples for analyses of tritium (and for other radionuclides present in sufficient concentrations).

Our FY 1990 transport research included continued monitoring of the RNM-2S well at Cambric, collection of samples from Cambric for technetium analysis, preparation for field sampling of colloids, and the drilling of UE3e#4. In this part of our research program, we seek to identify mechanisms that transport radioactive material from the immediate cavity region. At the Cambric site, we study the movement of several radionuclides with groundwater. At the UE3e#4 hole, we investigate the ways material moved to that location from the Sandreef site some 350 m away.

As mentioned earlier in this report, we monitor the radionuclide content of water pumped from the RNM-2S well at Cambric. We routinely measure the tritium and ^{85}Kr content of this water and boil

to dryness 108-1 samples in the search for ^{137}Cs . Even though ^{137}Cs concentration at the cavity has decreased, we have not detected ^{137}Cs at RNM-2S; cesium cations are known to sorb strongly on tuffaceous rock. We believe that cesium, which dissolves in the cavity region, moves only a short distance toward the pumped well before being sorbed on the tuffaceous rock. Tritium, which is in the form of tritiated water and is not sorbed on the rock, has moved away from the cavity with the groundwater flow. We have pumped about 91% of the original source term tritium from RNM-2S. Only about 41% of the source term ^{85}Kr was pumped out during the same time period.

Last year we measured ^{99}Tc in water from the Cambric cavity; this year we hope to measure ^{99}Tc in the water pumped from RNM-2S. Water samples from RNM-2S traced with ^{97}Tc were passed through a column containing an anion exchange resin. The technetium will be separated from the resin and counted by means of mass spectrometry. If these experiments are successful, we will have added another radionuclide to the list of those that can be used to trace the movement of material from the cavity to the pumped well at Cambric.

Colloid transport may be involved in the movement of radionuclides through the underground environment at the NTS. We have reviewed the literature on colloid collection techniques and have assembled the equipment. Field collection experiments will start at the Cambric site early in FY 1991.

Our major effort in transport studies during FY 1990 was the drilling of UE3e#4. We drilled this hole to discover the geologic and hydrologic conditions that enabled radionuclides to move more than 350 m from the Sandreef site to the Aleman site. The hole was drilled to a depth of 770 m; core and sidewall samples, as well as water samples, were collected. Three tubes, at three different depths, were placed in the hole to monitor water pressures and water content. We have issued an extensive report on the drilling at UE3e#4 and the analysis of samples from that site.

I. INTRODUCTION

This report describes the work accomplished during FY 1990 by Los Alamos National Laboratory personnel as participants in the Hydrology/Radionuclide Migration Project (HRMP). The US Department of Energy (DOE) supports this research on underground movement of radionuclides at the Nevada Test Site (NTS). We seek to measure the extent to which such movement occurs and to determine what mechanisms are involved. Organizations participating in 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 DOE coordinates activities within the HRMP; technical review of our work is provided by a steering committee of experts from relevant scientific disciplines.

HRMP research includes characterizing source terms, defining transport mechanisms, and identifying those aspects of the local and regional hydrology which may effect radionuclide movement. Los Alamos personnel are concerned with measuring the types and amounts of radionuclides and other hazardous materials (for example, heavy metals) present after an underground nuclear test and determining whether these materials move away from the immediate site of the explosion. The emphasis at LLNL is in these same areas, whereas our colleagues at USGS and DRI concentrate more on hydrologic work. Within Los Alamos, the divisions that contribute to HRMP research include the Isotope and Nuclear Chemistry Division, the Field Test Division, and the Earth and Environmental Sciences Division. The Yucca Mountain Project also studies the underground movement of radionuclides; some Los Alamos personnel contribute to that project as well as to the HRMP.

This report documents the HRMP research by Los Alamos personnel during FY 1990 and describes the resulting data; previous data are included as necessary to interpret recent results. Los Alamos research is highlighted in this report, but contributions from other participating organizations are identified. We have published a number of journal articles¹⁻³ and laboratory reports⁴⁻¹⁷ on various aspects of past research for this project.

II. PROGRAM MANAGEMENT

Geographical separation of HRMP's component organizations makes coordination of activities difficult. Assigning specific aspects of the project to each organization provides coherence in our program. During the past year, Los Alamos acted as the lead organization for research conducted in the Yucca Flats area of the NTS. In this capacity, we provided scientific oversight during the drilling of the hole UE3e#4 and assumed responsibility for drilling and sampling the chimney of hole U4ups2a.

Documentation resulting from these activities (drilling plans, reports, etc.) are listed in the Appendix of this report.

Los Alamos assists the DOE Nevada Operations Office by providing technical review of documents and preparing position papers on topics of importance to the HRMP. We also publish topical papers in the open literature. The Appendix lists the DOE and public papers prepared in FY 1990. The Appendix also includes a list of meetings attended by Los Alamos personnel as part of the programmatic effort.

III. SOURCE TERMS

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

The Cambric test conducted in 1965 has been studied by the HRMP for many years. The test took place below the water table in the tuffaceous alluvium of Frenchman Flat at the NTS. There is very little water movement at this location, and the radionuclides resulting from the test remained in the cavity region. The RNM-1 hole was drilled into the cavity at Cambric in 1974. After an auxiliary well, RNM-2S, was drilled 91 m away, we began pumping and continued pumping water at a rate of 2.3 m³/min. We monitor the concentrations of radionuclides in both cavity water and water from the pumped well.

Water samples collected annually from the RNM-1 well consist of 208-l samples in polyethylene-lined barrels, 2-l samples in air-tight stainless steel tubes, and additional samples of varying size collected in 20- to 0.5-l polyethylene bottles. We evaporate the large-volume samples and count the residues with a Ge detector to determine their gamma activity. The 2-l samples are processed on a vacuum line to separate ⁸⁵Kr, which is then measured by beta counting. We measure tritium by liquid scintillation counting. These sampling and counting protocols are described in more detail in Ref. 13.

The concentrations of tritium and ⁸⁵Kr in RNM-1 cavity water are given in Table I. In the last samples from the cavity, the levels of activity of both tritium and ⁸⁵Kr had fallen almost beyond the limits of our detection capability. The limits for tritium are approximately 1 pCi/ml of water and for ⁸⁵Kr, about 3×10^{-3} pCi/ml of water. Apparently, as water was pumped from the RNM-2S well, krypton and tritium have moved away from the cavity at approximately the same rate. That is, the atom ratio of ⁸⁵Kr to tritium has been roughly constant (save for one measurement in 1977) over 15 years. This should be expected because both these radionuclides are mobile, and neither interacts strongly with the alluvium through which it is moving. It is interesting that the ratio of ⁸⁵Kr to tritium in cavity water

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

Zone	Date	Volume Pumped ^a (10^6 m^3)	Concentration ^b Tritium (mCi/ml)	Concentration ^{85}Kr (mCi/ml)	^{85}Kr /Tritium	
					Atom ratio ^c ($\times 10^4$)	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	$\sim 1 \times 10^{-6}$	$\sim 4 \times 10^{-9}$	—	—
Zone IV + V	05/16/90	15.9	$\sim 1 \times 10^{-6}$	$\sim 2 \times 10^{-9}$	—	—

^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}).

at Cambric was initially higher than the calculated source term value, and it remained higher throughout the pumping.

The concentrations of ^{90}Sr and ^{137}Cs in the RNM-1 water are shown in Table II. The concentration of ^{90}Sr has been near our limits of detection for several years; recent fluctuations of the values reported in the table reflect the uncertainties in these measurements. In contrast, we can detect ^{137}Cs at concentrations several orders of magnitude lower than those current at the RNM-1 cavity. The cumulative data in Table II indicate that strontium and cesium, like tritium and krypton, have decreased considerably in the cavity region during the period of pumping at the satellite well. However, the rate of decrease for these two elements is lower than that for tritium, so the atom ratios of $^{90}\text{Sr}/\text{T}$ and $^{137}\text{Cs}/\text{T}$ have grown progressively larger. We believe that strontium and cesium initially trapped in melt glass at the bottom of the cavity are dissolving (leaching) and that ions of these elements sorbed on the rock surfaces in the cavity/chimney complex are desorbing. The combination of these processes tends to raise the concentrations of these elements even as pumping tends to lower them. In the future, ^{137}Cs appears to be the only radionuclide we can expect to measure with reasonable precision in the Cambric cavity water.

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

Immediately following a nuclear detonation underground, a cavity forms where the device was originally placed. Vaporized material—rock, material associated with the device, and products of the fission and fusion processes—fills the cavity initially. As this vapor cools and condenses, materials collect on the walls and floor of the cavity. At this point, some separation occurs between refractory elements and more volatile ones. This separation is not complete; refractory elements like zirconium are present in locations other than in the puddle glass, which forms on the floor of the cavity, and volatile elements like iodine are present even in the puddle glass. If the cavity collapses (as it generally does), materials initially condensed on the cavity ceiling fall to the cavity floor, thus reversing some of the spatial separation between volatile and refractory materials. These processes are discussed in some detail in Ref. 18.

During FY 1991, Los Alamos researchers examined materials collected in postshot drillbacks from shots of various yields in various media and determined the extent of separation of volatile and refractory radionuclides. We also studied the leaching characteristics of these radionuclides in comparison with their location in the postshot cavity/chimney complex. This research is important in characterization of the source terms for possible radionuclide migration away from event sites. Because many tests are conducted above the standing water level (SWL), the resultant puddle glass may end up below the water level, and radioactive material condensed in the chimney may extend into the vadose

Table II. Tritium, ^{90}Sr , and ^{137}Cs Concentrations in RNM-1 Water

Zone	Date Collected	Volume Pumped ^a (10^6 m^3)	Concentration ^b			Atom Ratio ^c ($\times 10^4$)	
			Tritium ($\mu\text{Ci/ml}$)	^{90}Sr ($\mu\text{Ci/ml}$)	^{137}Cs ($\mu\text{Ci/ml}$)	$^{90}\text{Sr}/\text{T}$	$^{137}\text{Cs}/\text{T}$
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	$\sim 1 \times 10^{-6}$	2×10^{-8}	1.2×10^{-8}	~ 500	~ 300
Zone IV + V	05/16/90	15.9	$\sim 1 \times 10^{-6}$	—	9.4×10^{-9}	—	~ 200

^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 $^{137}\text{Cs} = 1 \mu\text{Ci}$.

zone. In our FY 1989 report¹⁷, we described the methodology used in these experiments and the calibration procedures for our counters. We reported initial studies of debris from two underground tests that resulted in spatial differentiation of volatile and refractory materials. Some differences in dissolution rates between the volatile and refractory materials were observed. We have continued this research with debris from three recent underground nuclear tests identified here as Shot 1, Shot 2, and Shot 3.

1. Shot 1

This test was conducted 16 m below the SWL in zeolitized, ash-flow tuff. The water content of the tuff was 18.3% by weight. The porosity was 38% by volume and the saturation was 87% by volume. Samples were obtained from one hole at 556-m vertical depth and from another hole at depths of 557, 553, 546, and 544 m. These samples were finely pulverized with a ball mill; however, a portion of the sample from 544 m was retained as "high-graded" material depleted in non-debris components. The samples were counted with a Ge detector and the concentrations of gamma-emitting radionuclides were determined by analysis codes (GAMANAL and CLSQ). The data are shown in Table III.

The concentrations of the elements zirconium, cerium, and neodymium decrease as the sampling moves away from the puddle region. In contrast, the concentrations of ruthenium, antimony, and iodine are somewhat larger at locations higher in the hole. These data generally conform to our expectations, which are based on the relative volatilities of these elements. However, even the most refractory elements can be found at distance from the puddle and even the most volatile elements are present in the puddle region. After being counted, the samples were placed in centrifuge tube with water from the J-13 well at the NTS (water : rock ratio of 20 ml : 1 g) and shaken at 70°C for 16 days. The phases were then separated by filtering through 50-nm Nuclepore filters and the leachates were analyzed for their radionuclide content by both the Ge detector and the analysis codes. The results of the leaching experiment are shown in Table IV. The top set of numbers provide the concentrations of each radionuclide per ml of water after 16 days of leaching. The bottom set of numbers (in bold print) is the concentration in solution divided by the original concentration of the solid phase; that is, this number is an indication of the relative amount of leaching that took place for each measured radionuclide. Note that only a few radionuclides in the leachate are of sufficient concentration to be detected.

Table III. Concentration of Radionuclides (atoms/g) vs Depth of Sample — Shot 1

Nuclide	556 m	557 m	553 m	546 m	544 m	544m ^a
⁹⁵ Zr	7.1 x 10 ¹²	7.5 x 10 ¹²	1.4 x 10 ¹²	8.4 x 10 ¹⁰		5.0 x 10 ⁸
¹⁴⁴ Ce	6.4 x 10 ¹²	6.3 x 10 ¹²	1.2 x 10 ¹²			
¹⁴¹ Ce	4.9 x 10 ¹²	4.2 x 10 ¹²	8.8 x 10 ¹¹	8.4 x 10 ¹¹		3.7 x 10 ⁹
¹⁴⁰ Ba	3.1 x 10 ¹²	2.4 x 10 ¹²	5.6 x 10 ¹¹	3.0 x 10 ¹²		1.7 x 10 ¹⁰
¹⁴⁷ Nd	2.4 x 10 ¹²	2.7 x 10 ¹²	4.9 x 10 ¹¹			
²³⁷ U	1.1 x 10 ¹²	1.1 x 10 ¹²	2.1 x 10 ¹¹			
¹³¹ I	9.7 x 10 ¹¹	8.4 x 10 ¹¹	2.7 x 10 ¹¹	1.0 x 10 ¹²	3.5 x 10 ⁹	2.6 x 10 ¹⁰
¹⁰³ Ru	8.2 x 10 ¹¹	1.1 x 10 ¹²	2.6 x 10 ¹¹	5.0 x 10 ¹¹	1.3 x 10 ⁸	3.6 x 10 ¹⁰
¹²⁴ Sb	1.7 x 10 ¹¹	1.5 x 10 ¹¹	7.6 x 10 ¹⁰	2.2 x 10 ¹¹		5.4 x 10 ⁹

^a"High-graded" to remove nondebris material.

Table IV. Concentration of Radionuclides (atoms/ml) in Leachate — Shot 1

Nuclide	$\frac{\text{atoms / ml in leachate}}{\text{atoms/g in solid}} \quad \text{after 16 days at 70}^\circ\text{C}$					
	556 m	557 m	553 m	546 m	544 m	544m ^a
¹⁰³ Ru	8.8 x 10 ⁷ 1.1 x 10 ⁻⁴	3.7 x 10 ⁷ 3.4 x 10 ⁻⁵	4.6 x 10 ⁷ 1.8 x 10 ⁻⁴	2.3 x 10 ⁷ 4.6 x 10 ⁻⁵	3.0 x 10 ⁶ 2.3 x 10 ⁻⁵	
¹²⁴ Sb	2.7 x 10 ⁸ 1.6 x 10 ⁻³	2.0 x 10 ⁸ 1.3 x 10 ⁻³	6.0 x 10 ⁷ 7.9 x 10 ⁻⁴	1.0 x 10 ⁸ 4.6 x 10 ⁻⁴		2.0 x 10 ⁷ 3.7 x 10 ⁻³
²³⁷ U	9.2 x 10 ⁸ 8.4 x 10 ⁻⁴	8.9 x 10 ⁸ 8.1 x 10 ⁻⁴				
¹³¹ I	4.6 x 10 ⁹ 4.7 x 10 ⁻³	6.9 x 10 ⁹ 8.2 x 10 ⁻³	3.1 x 10 ⁹ 1.1 x 10 ⁻²	1.2 x 10 ⁹ 1.2 x 10 ⁻³	6.0 x 10 ⁸ 1.7 x 10 ⁻¹	2.3 x 10 ⁸ 8.8 x 10 ⁻³

^a"High-graded" to remove nondebris material.

2. Shot 2

This test was conducted in tunnel bed tuffs 331 m above the SWL. The water content of the tuff was 12% by weight; the porosity was 42% by volume and the saturation was 46% by volume. The samples we analyzed were taken from three different depths in the drillback hole, and had been ground with a ball mill. These samples were counted and analyzed for gamma-emitting radionuclides, and the results are shown in Table V. Subsequent to the initial counting, the samples were shaken with J-13 water (20 ml of water to 1 g of rock) at 70°C for 26 days. The leachate was filtered through 50-nm Nuclepore filters and analyzed on the Ge counter. Data from the leaching experiment are shown in Table VI.

3. Shot 3

The environment for this test was similar to that for Shot 2. The working point was in tunnel bed tuffs 327 m above the water table. The water content of the tuff was 14% by weight, the porosity was 43% by volume, and the saturation was 52% by volume. Three samples, ground with a ball mill, were taken from varying heights in the drillback hole. After the samples were counted, they were leached with J-13 water (20 ml of water to 1 g of rock) at 70°C for 40 days. The leachate was filtered and analyzed as before. Data from these experiments are shown in Tables VII and VIII.

Table V. Concentration of Radionuclides (atmos/g) vs Depth of Sample - Shot 2			
Nuclide	262 m	257 m	251 m
⁹⁹ Mo	5.4 x 10 ¹²	2.5 x 10 ¹²	2.4 x 10 ¹¹
⁹⁵ Zr	4.7 x 10 ¹²	2.4 x 10 ¹²	9.2 x 10 ¹⁰
¹⁴³ Ce	3.7 x 10 ¹²	2.8 x 10 ¹²	
²³⁹ Np	3.1 x 10 ¹²	2.6 x 10 ¹²	
¹⁴¹ Ce	2.5 x 10 ¹²	4.4 x 10 ¹²	2.1 x 10 ¹⁰
¹⁰³ Ru	2.3 x 10 ¹²	1.8 x 10 ¹²	1.0 x 10 ¹²
¹⁴⁷ Nd	2.0 x 10 ¹²	1.1 x 10 ¹²	3.4 x 10 ¹⁰
¹⁴⁰ Ba	1.6 x 10 ¹²	4.0 x 10 ¹²	3.9 x 10 ¹¹
¹³¹ I	4.5 x 10 ¹¹	1.4 x 10 ¹²	5.5 x 10 ¹¹
¹³² Te	3.8 x 10 ¹¹	1.2 x 10 ¹²	7.2 x 10 ¹¹
⁵⁴ Mn	1.9 x 10 ¹¹	1.3 x 10 ¹¹	6.3 x 10 ⁹
¹²² Sb	1.9 x 10 ¹¹	7.3 x 10 ¹¹	1.5 x 10 ¹¹
²³⁷ U	1.3 x 10 ¹¹	1.2 x 10 ¹¹	
¹²⁴ Sb	1.1 x 10 ¹¹	4.5 x 10 ¹¹	1.0 x 10 ¹¹
¹²⁷ Sb	8.9 x 10 ¹⁰	3.0 x 10 ¹¹	4.6 x 10 ¹⁰
¹³⁶ Cs	3.9 x 10 ¹⁰	1.0 x 10 ¹¹	1.5 x 10 ¹⁰

**Table VI. Concentration of Radionuclides (atmos/ml)
in Leachate — Shot 2**

Nuclide	after 26 days at 70°C		
	<u>atoms/ml in leachate</u> atoms/g in solid		
	262 m	257 m	251 m
⁹⁵ Zr	7.2 x 10 ⁶ 1.5 x 10 ⁻⁶	1.7 x 10 ⁸ 7.4 x 10 ⁻⁵	1.9 x 10 ⁹ 2.1 x 10 ⁻³
¹⁰³ Ru		1.3 x 10 ⁸ 7.3 x 10 ⁻⁵	4.6 x 10 ⁷ 4.6 x 10 ⁻⁵
¹⁴⁷ Nd	1.2 x 10 ⁷ 6.0 x 10 ⁻⁶	2.1 x 10 ⁷ 2.0 x 10 ⁻⁵	9.0 x 10 ⁶ 2.7 x 10 ⁻⁴
¹²⁴ Sb	1.1 x 10 ⁷ 1.0 x 10 ⁻⁴	7.1 x 10 ⁸ 1.6 x 10 ⁻³	6.9 x 10 ⁸ 6.8 x 10 ⁻³
¹³¹ I	7.7 x 10 ⁸ 1.7 x 10 ⁻³	5.7 x 10 ⁹ 4.0 x 10 ⁻³	9.4 x 10 ⁹ 1.7 x 10 ⁻²

**Table VII. Concentration of Radionuclides (atmos/g)
vs Depth of Sample — Shot 3**

Nuclide	229 m	216 m	209 m
⁹⁹ Mo	1.5 x 10 ¹³	3.3 x 10 ¹¹	4.8 x 10 ¹⁰
²³⁹ Np	1.4 x 10 ¹³	3.6 x 10 ¹¹	5.5 x 10 ¹⁰
⁹⁵ Zr	1.3 x 10 ¹³	2.8 x 10 ¹¹	4.0 x 10 ¹⁰
¹⁴³ Ce	1.0 x 10 ¹³	2.4 x 10 ¹¹	3.6 x 10 ¹⁰
¹⁴⁴ Ce	8.9 x 10 ¹²	2.3 x 10 ¹¹	3.0 x 10 ¹⁰
¹⁰³ Ru	7.5 x 10 ¹²	2.1 x 10 ¹¹	3.4 x 10 ¹⁰
¹⁴¹ Ce	6.7 x 10 ¹²	1.9 x 10 ¹¹	2.9 x 10 ¹⁰
¹⁴⁷ Nd	5.3 x 10 ¹²	1.2 x 10 ¹¹	1.8 x 10 ¹⁰
¹³⁴ Cs	4.5 x 10 ¹²	1.8 x 10 ¹¹	2.9 x 10 ¹⁰
¹⁴⁰ Ba	4.1 x 10 ¹²	1.3 x 10 ¹¹	2.1 x 10 ¹⁰
¹³¹ I	1.6 x 10 ¹²	5.9 x 10 ¹⁰	1.1 x 10 ¹⁰
¹³² Te	1.3 x 10 ¹²	4.4 x 10 ¹⁰	1.0 x 10 ¹⁰
¹²² Sb	6.0 x 10 ¹¹	2.2 x 10 ¹⁰	3.5 x 10 ⁹
²³⁷ U	3.8 x 10 ¹¹	3.7 x 10 ¹⁰	5.2 x 10 ⁹
¹²⁷ Sb	3.4 x 10 ¹¹	1.4 x 10 ¹⁰	2.6 x 10 ⁹
¹²⁴ Sb	3.0 x 10 ¹¹	3.9 x 10 ⁹	1.8 x 10 ⁹
⁵⁴ Mn	4.2 x 10 ¹¹	1.0 x 10 ¹⁰	
¹³⁶ Cs	9.7 x 10 ¹⁰	3.2 x 10 ⁹	5.4 x 10 ⁸
⁵⁹ Fe	4.0 x 10 ¹⁰	2.2 x 10 ⁹	

**Table VIII. Concentrations of Radionuclides (atmos/g)
in Leachate — Shot 3**

Nuclide	<u>atoms/ml in leachate</u> after 40 days at 70°C		
	atoms/g in solid		
	229 m	216 m	209 m
^{141}Ce	1.0×10^7 1.5×10^{-6}	1.8×10^6 9.5×10^{-6}	1.6×10^6 5.5×10^{-5}
^{103}Ru	1.4×10^7 1.9×10^{-6}	3.8×10^5 1.8×10^{-6}	2.8×10^5 8.2×10^{-6}
^{140}Ba	2.2×10^7 5.4×10^{-6}	6.5×10^6 5.0×10^{-5}	
^{134}Cs	3.0×10^8 6.7×10^{-5}	9.6×10^6 5.3×10^{-5}	3.7×10^7 1.3×10^{-3}
^{124}Sb	5.2×10^7 1.7×10^{-4}		2.6×10^6 1.4×10^{-3}
^{131}I	7.8×10^8 4.9×10^{-4}	1.1×10^7 1.9×10^{-4}	3.1×10^7 2.8×10^{-3}

4. Spatial Separation of Refractory and Volatile Elements

In Table IX we have compared the distribution of radionuclides in debris from the three shots. For each shot, we tabulated the ratio of the radionuclide concentration high in the cavity to radionuclide concentration close to the bottom. The pattern of these ratios is similar for all three shots; that is, the relative amounts of volatile elements like iodine, tellurium, and antimony are greater with distance from the cavity bottom than are similar ratios for refractory elements like zirconium, molybdenum, neptunium, and cerium. The variations between refractory and volatile elements is similar for Shot 1 and Shot 2, even though these tests were conducted in different geologic environments. However, Shot 3, in an environment essentially identical to that of Shot 2, showed much less separation of refractory and volatile elements. Again we emphasize that the separation is not complete; samples from high in the drillback hole contain some refractory material and samples from the cavity bottom contain some volatile material. The number of radionuclides we can identify depends to a considerable extent on how soon the debris is available for counting. Radionuclides with half-lives of a few days—like ^{99}Mo , ^{239}Np , ^{143}Ce , ^{132}Te , and ^{237}U —soon decay to concentrations below our detection limits. We will continue to examine new distribution patterns as we collect data from more tests.

**Table IX. Relative Concentrations of Radionuclides:
At Distance from the Bottom of the Cavity
vs Close to the Bottom of the Cavity**

Radionuclide	Shot 1 ($\frac{456\text{m}}{546\text{m}}$)	Shot 2 ($\frac{251\text{m}}{262\text{m}}$)	Shot 3 ($\frac{209\text{m}}{229\text{m}}$)
^{95}Zr	1.2×10^{-2}	2.0×10^{-2}	3.1×10^{-3}
^{99}Mo		4.4×10^{-2}	3.2×10^{-3}
Np			3.9×10^{-3}
^{143}Ce			3.6×10^{-3}
^{141}Ce	1.7×10^{-1}	8.4×10^{-3}	4.3×10^{-3}
^{103}Ru	6.1×10^{-1}	4.3×10^{-1}	4.5×10^{-3}
^{147}Nd		1.7×10^{-2}	3.4×10^{-3}
^{140}Ba	9.7×10^{-1}	2.4×10^{-1}	5.1×10^{-3}
^{136}Cs		3.8×10^{-1}	5.6×10^{-3}
^{134}Cs			6.4×10^{-3}
^{122}Sb		7.9×10^{-1}	5.8×10^{-3}
^{124}Sb	1.3	9.1×10^{-1}	6.0×10^{-3}
^{127}Sb		5.2×10^{-1}	7.6×10^{-3}
^{131}I	1.0	1.2	6.9×10^{-3}
^{132}Te		1.9	7.7×10^{-3}
^{237}U			1.4×10^{-2}

5. Leaching of Shot Debris

The results of only three experiments and the relatively few radionuclides at detectable concentrations in the leachates make it hard to generalize. Nevertheless, the data do show some interesting trends that we hope to confirm with more experiments. In Tables IV, VI, and VIII, the bold print numbers for each radionuclide (with few exceptions) increase in magnitude reading from left to right. The leachability of each radionuclide appears to increase as the point of sampling moves upward in the hole. Thus, the ^{103}Ru that was associated with puddle glass is less leachable than the ^{103}Ru that was condensed some distance away from the puddle. The same pattern is reflected in the leachability of the volatile radionuclide ^{131}I . Part of this difference in leachability may be due to different quantities of material being deposited in different regions of the cavity: thin films at the top of the cavity and thick layers at the bottom. Presumably the former would leach more rapidly than the latter. Such bulk effects should be partially negated by the grinding which each sample underwent before it was used in the leaching studies. If our hypothesis holds true, the regions surrounding a cavity bottom will prove a more significant source for dissolved radionuclides than the puddle glass at the cavity bottom. More data are needed before we can judge the validity of this interpretation.

C. SURVEY OF HAZARDOUS MATERIALS USED IN NUCLEAR TESTING (E. A. Bryant and J. Fabryka-Martin)

1. Introduction

Concerns about the environmental impact of "weapons complex" activities motivated us to review underground testing practices at the Nevada Test Site (NTS) and to examine the nature and quantity of the "hazardous" materials used. This information can be used in our evaluation of potential environmental impacts resulting from migration of hazardous waste material away from the sites of the nuclear explosions.

This report summarizes the results of our review in four sections: (1) a summary of the history of underground nuclear testing, (2) a description of current test practices, (3) a summary of sources for test material information, and (4) the results from a trial inventory for a specific test. The first section reviews testing by both Los Alamos National Laboratory (LANL) and Lawrence Livermore National Laboratory (LLNL), but the other sections relate only to LANL testing. A brief introduction to relevant hazardous material definitions appears at the end of this section.

2. Brief Summary of Underground Testing at the Nevada Test Site

A brief history of the underground testing at the NTS allows us to gain a perspective on the nature and quantity of hazardous materials used. Because many of the same materials are used for each nuclear test, we need to know how many tests have taken place. Larger quantities of materials are needed for larger explosions and the inventory of hazardous waste may be proportional to total test yield rather than to the number of tests.

Also, the energy released by the test determines the degree of dilution of hazardous materials through mixing with melted and volatilized geologic materials. Documenting both the number and total yield of the nuclear tests aids in this examination.

From the beginning of underground testing of weapons at the NTS in 1957 through February 1990, a total of 616 publicly announced underground nuclear tests have occurred. (All subsequent references to tests in this document are limited to publicly announced tests.)

The number of tests conducted near or below the water table is directly related to the hazardous waste migration in ground water. There have been 172 tests for which the shot points or lower cavity edge lay below the SWL (Table X). Of these tests, 52% were in Yucca Flats and 39% in Pahute Mesa.

As of February 1990, the total fission yield for NTS events conducted below the water table or with a cavity edge extending below the water table was about 28 megatons (Mt). This measurement is a factor of 4 larger than the value estimated by Borg¹⁸ for the period ending June 1975 (Table XI). Expressed in terms of yield, 68% of the "ground-water-accessible" radionuclide inventory was deposited in Pahute Mesa.

3. Weapons Test Practices

The following subsections describe steps required to conduct a nuclear test. In each case, we emphasize the introduction and/or redistribution of materials in the vicinity of the test.

Table X. Distribution of Announced Underground Tests at the NTS (1957 - 1990)

	Shot Point Below SWL	Lower Cavity Below SWL	Lower Cavity <75 m Above SWL	Cavity >75 m Above SWL	Total Number of Events
Pahute Mesa	33	33	6	5	77
South Yucca Flat	10	4	23	140	177
North Yucca Flat	67	23	32	189	311
Frenchman Flat	1	1	2	0	4
Rainier Mesa	0	0	0	47	47
TOTAL NUMBER	111	61	63	381	616

Definition of geographical units: Pahute Mesa = areas 18, 19, 20; South Yucca Flat = areas 1, 3, 6, 11; North Yucca Flat = areas 2, 4, 7, 8, 9, 10, 15; Frenchman Flat = area 5; Rainier Mesa = area 12.

**TABLE XI. Geographical Distribution of Test Yields
When the Depth of Burial or Cavity Edge
Extended Below the Water Table**

	Borg Report (as of 6/75)	Updated Estimate (as of 2/90)
Pahute Mesa	4 Mt	19 Mt
Yucca Flat	3 Mt	9 Mt
Frenchman Flat	<0.01 Mt	<0.01 Mt
SUBTOTAL	7 Mt	28 Mt

a. Drilling and Hole Preparation

Emplacement holes, 2.44 m in diameter, are drilled with polymer or detergent and water by a dual-string reverse circulation method. This dual-string reverse circulation method has replaced the conventional circulation method that used bentonite or sepiolite mud.

Los Alamos Group HSE-1 collects samples of drilling fluid daily to analyze for gross radioactivity. Group HSE-1 also collects water samples for ^3H assay.

A steel casing, extending 9 to 30 m below the surface, is installed in the emplacement holes. If the test point is below the SWL, a liner is installed in the bottom of the emplacement hole and the hole is blown dry; otherwise, no liner is installed. Cement grout is placed around the casing and liner.

b. Nuclear Device and Rack

Each test uses a test rack, a steel structure that supports the nuclear device and the instruments that measure the results. Typically more than 30 m in height, the racks include from 2 to 20 line-of-sight pipes. Each of these pipes has a window whose composition is compatible with the desired measurement (aluminum, beryllium, etc.). The rack sits on top of a steel canister, often lined with a mixture of boron and polyethylene, that contains the device.

The racks are fabricated and assembled under the direction of the LANL Test Engineering Group, J-7. Detectors are designed by Groups P-14 (Fast Transient Plasma Measurements Group) and P-15 (Neutron Measurements Group) and fabricated by EG&G; some specific parts may be provided by the experimenter. Occasionally, experiments or objects are included on the rack for other groups, such as Group J-8 (for timing and firing). These additional test items are generally coordinated through J and/or P Division.

Large quantities of polyethylene are used on the racks. Other organic materials used include polyvinyl chloride (PVC), Teflon, polystyrene, phenolic, and neoprene. Potentially hazardous organic chemicals include complex fluorescing compounds (liquid scintillator) and laser dyes used as part of some detector packages.

Among the inorganic hazardous materials, lead is used extensively. Typically, tens of tons are used for shielding in both the canister and rack. Copper is also used: always for wiring but sometimes for other purposes such as shielding. Beryllium, nickel, and zinc may be present in small quantities in detector packages (~100 g). Arsenic, chromium, cadmium, osmium, and thallium have been used in rare

instances. Other commonly used metals include tungsten, tantalum, stainless steel (iron, chromium, and nickel), and aluminum.

Each test device contains nuclear materials such as uranium, plutonium, tritium, lithium, and structural materials such as steel, aluminum, beryllium, gold, etc. Radiochemical detectors (for example, yttrium, zirconium, thulium, lutetium) and tracers (isotopes of uranium, plutonium, americium, or curium) are also used. The nature and quantity of these materials cannot be reported in an unclassified document. Generally, the detectors and tracers are used in less than 100-g quantities.

c. Back-Fill

Magnetite (major ingredient Fe_3O_4) powder is poured downhole to cover the sides and top of the rack. This naturally occurring material contains thorium and other impurities such as heavy rare earths.

Stemming materials in the emplacement hole are used to prevent the upward escape of radioactivity from the device. The stemming consists of layers of pea gravel alternating with layers of "fines" (fine gravel)—native materials from the NTS shaker plant—and two or more special plugs—made of either two-part epoxy (TPE) or coal-tar epoxy (CTE)—placed well outside the melt zone. These plugs remain intact following the test.

d. Detonation

Initially, the explosion creates an approximately spherical cavity filled with gases formed by the atomization and vaporization of materials from the explosive device and its immediate surroundings. The molten cavity walls subsequently flow down to form a puddle that later freezes to form glassy material. The rock above the cavity eventually falls down to fill the cavity with rubble; this chimney-forming process may continue all the way up to the surface to form a crater, or it may stop at some intermediate point. The vaporized material either condenses and incorporates into molten rock or it escapes into the chimney rubble, where it may condense on solid rock. Volatile elements/materials tend to be enriched in the rubble zone; whereas refractory materials tend to remain in the puddle glass.

The melt zone created by the nuclear test incorporates a mass (expressed in tons) of the same order of magnitude as the device yield (expressed in tons); thus the zone would extend beyond the top of a 30-m rack if the yield is about 100 kt or more. In a test with a significant nuclear energy release, the entire device is atomized and mixed with a relatively large quantity of rock.

e. Re-Entry

Drill-back for diagnostic sample recovery involves drilling a hole, about 25 cm in diameter, at an angle that is directed to intercept the test debris puddle near its center. A profile of the radioactive material distribution along the length of the hole is measured with a downhole Geiger counter, and then samples of the puddle glass are collected with a sidewall core sampler. The drilling procedure uses drilling mud with various additives. A significant fraction of the mud is generally lost downhole into the more-or-less open structure of the rubble created by the test. Whereas LLNL uses "air-foam" for the upper part of the drill-back hole, LANL almost always uses mud for the entire hole.

Drilling mud is prepared by pumping water from a water-supply well to a surface impoundment and then to the REECO Mud Plant. Water for drilling in the flats is obtained from wells C, C-1, and C-4; wells A and UE1r were used in the past but were shut down a few years ago. On Pahute Mesa, local water wells and a portable mud plant are used. An inventory of drilling-mud components stocked in the REECO Mud Plant appears in Table XII; it was assembled by Barbara Hargis (formerly of Group HSE-8) for Dale Engstrom (Group J-6) in 1989.

Hazardous chemicals included in Table XII are chrome (in Raykrome 400), ethylenediamine (in Soda Ash), and paraformaldehyde (in Magcocide and My-Lo-Jel preservative). The unofficial memo to Dale Engstrom notes that other chemicals have been used in formulating the drilling mud, but that they are ordered only when necessary and are not stocked.

Drilling fluids are sampled at irregular intervals by several groups. D. Henderson of Group HSE-1 monitors drilling fluids for radioactivity during drilling of new emplacement holes. During 1989, Tony Grieggs of Group HSE-8 sampled mud from three drill-backs (UE19ay, UE7bk, and U4s) to determine whether the mud should be classified as hazardous waste because of its heavy metal or organic contents. Using US Environmental Protection Agency (EPA) approved procedures, Group HSE-9 analyzed the mud samples for eight toxic metals and for semi- and volatile organics. None of the muds exceeded EPA toxicity limits; in fact, the levels measured less than the EPA limits by more than an order of magnitude in every case. Relatively few organics were noted; T. Grieggs suggests that the observed organics may be traceable either to contamination in the laboratory or to petroleum breakdown products. More sampling is tentatively planned for 1990.

TABLE XII. Materials Used in Drilling at the NTS^a

Trade Name	Chemical Material
INORGANIC MATERIALS	
Hydrogel, Big Horn or Envirogel	Sodium montmorillonite, Western Bentonite
Thermogel	Sepiolite
Caustic soda	NaOH
"Potash"	97% KCl
ORGANIC MATERIALS	
Cypan	Sodium polyacrylate (polymer)
Thatcher Foamer TF	Surfactant TF foamer containing isopropanol
Magconol	Alcohol
Cydril 4000 Flocculant	Anionic polyacrylamide
Soda Ash	Contains theophylline, ethylenediamine, carbonic acid disodium salt
Guar Gum	Galacto-mannans (C ₆ H ₁₀ O ₅) _n
Rapid Mud	Liquid anionic polyelectrolyte (organic)
Raykrome 400	Chrome lignosulfonate, contains 4% Cr
My-Lo-Jel	Pre-gelatinized starch
Polysal	Modified starch (drilling fluid compound to reduce fluid loss)
Magcocide	91% paraformaldehyde (EPA hazardous chemical)
My-Lo-Jel preservative	95% paraformaldehyde (EPA hazardous chemical)
COMPOSITION UNKNOWN	
Magco Foam Check	Proprietary mixture

^aCompiled by B. Hargis (Group HSE-8) for D. Engstrom (Group J-6).

4. Records of Material Used in Nuclear Tests

Information concerning the nature and quantity of materials used in any particular underground nuclear test must be obtained from a number of sources. Generally, records for each operation are kept by only one organization.

a. Drilling Records (Field Engineering, Group J-6, Dale Engstrom)

Drilling is carried out by REEC Co under the direction of Group J-6; REEC Co prepares a daily drilling report on each hole. Phyllis Rashki of REEC Co also maintains drilling records that concern hazardous waste. Fenix and Scisson, the engineering contractor for drilling, compiles the history and data for each hole; this history is then sent to Group J-6, the Department of Energy at NTS, Jack House, Program Manager for Containment (EES-DO), and Group EES-3 (Containment). These Group J-6 records are sent to LANL Central Storage after about 1 year.

b. Rack and Canister Materials

The materials contained in each rack are not listed in a data base; however, the Coordinator designated for each test can provide an overall picture of the various experiments on the rack. Information on rack contents can generally be obtained from the four different sources listed here.

- (1) Rack drawings, maintained in the Group J-7 Office, Building SM-216. These drawings include details of the rack structure, shielding, and each experiment—but generally not the detectors (measurement devices)—within each experiment.
- (2) Detector handbook, maintained by EG&G. This handbook documents detectors and their components. Such information can alternatively be found in P-Division files. All detectors are built by EG&G under an extensive QA plan.
- (3) Group P-14 or Group P-15 Progress Reports or "shot reports" prepared by an experimenter for a particular experiment. These reports describe custom parts supplied by individual experimenters. Shot reports are distributed at the discretion of the experimenter but generally go to X Division Office (X-DO), P-DO, J-DO, and Groups X-2, X-4, P-14, and P-15.
- (4) Laboratory notebooks of individual experimenters. In some instances, the materials in a particular experiment may be known only to the experimenter and can be found only in his laboratory notebook. This practice was probably more common in the past.

c. Device Materials

Nuclear devices may contain or produce radioactive isotopes of the actinides (uranium, neptunium, plutonium, and americium in particular), tritium, and fission products. In addition, various metals may be incorporated as structural materials or as neutron flux monitors. The nature and quantity of ingoing materials, well documented in Classified data bases, will not be discussed further in this Unclassified report.

d. Relevant Data Bases

Information about the tests conducted at the NTS can be obtained from the sources listed here.

- (1) The shot book maintained in INC-Division vault. Starting with Trinity in 1945 and including shots that occurred outside the NTS, this book provides a chronological listing of all US tests. This book contains shot number, name, laboratory, date and time, emplacement area and hole, and location (for example, underground depth and type of rock).
- (2) Containment reports for individual tests. Currently prepared by EES Division, these reports are available for all LANL tests. Similar reports are available for a few LLNL tests. These reports include information about:
 - hole construction and drilling logs; x-ray analyses of drill cuttings; materials used for drilling and grouting;
 - nearby holes;
 - diagnostic line-of-sight assemblies and plugs for each assembly (for example, plug compositions include aluminum, lead, copper, bromine, iron, tungsten, PVC, and polyethylene (CH₂));
 - emplacement hole design plans for containment (back-fill material and plugs);
 - site geology, nearby faults, stratigraphy, lithology, geologic cross-sections; and
 - event yield, diagnostics system, working point parameters (such as porosity, geology, and depth to water table), containment design (for example, thickness and composition of layers and plugs).
- (3) COEDS (COmmon Event Data System) (Nancy Marusek, Group EES-5 and Project Leader, Don Shirk, Group X-5). This data base includes data for both LANL and LLNL tests. Several LANL divisions, including INC, X, J, WX, and EES, maintain tables within this system. Types of data include depth of burial, water level, measured and estimated cavity radius, and test yield.

- (4) GEODES (GEOlogic Data Evaluation System) (Nancy Marusek, Group EES-5). The GEODES data base contains downhole information on LANL tests, including lithologic logs, location, and SWL.
- (5) Pre-shot data base (Tim Benjamin, Group INC-7). This data base, containing SECRET RESTRICTED DATA, is maintained by Groups INC-7 and 11 (Scott Bowen and Zita Svitra) on the INC DP2 Classified VAX. Although most LANL tests are included, many LLNL tests are missing. The data base contains material summaries for the nuclear devices. It includes all major elements, plus trace elements that could be significant in diagnostic interpretations. Isotopics of fissile material and elemental compositions taken from design engineering drawings from WX Division are also included. Analyses of isotopic signatures and assays of impurities are based on information from CLS or INC Divisions, Rocky Flats, or other sources.
- (6) Post-shot data base (Tim Benjamin, Group INC-7). This data base is maintained by Groups INC-7 and INC-11 on the INC DP2 Classified VAX. It contains concentration and isotopic data from analyses of drill-core samples for fission products, actinides, and radiochemical detectors.

5. Trial Inventory for the Amarillo Test

We tested the accessibility and quality of data available to estimate a complete downhole inventory by compiling available information for Amarillo, which was fired June 27, 1989, in emplacement hole UE19ay. This test was selected because its yield was fairly typical; it was conducted below the water table; and it was one of the three tests sampled by Tony Grieggs of Group HSE-1 to evaluate drill-back mud as a potential hazardous waste.

The following sections discuss the data source and the nature and amounts of hazardous chemical wastes emplaced downhole.

a. Emplacement Hole Drilling

A copy of the Fenix & Scisson, Inc., "Hole History Data" was provided by Group J-6. This document summarizes the activities for each day of hole preparation and the amounts of Rapid Mud and cement used. No hazardous materials were identified.

b. Rack

A complete set of rack drawings was provided by Group J-7. These drawings identified materials but generally not the amounts used (except for lead); these amounts could be deduced from volume and density. The following materials were used at one or more locations: lead, tungsten, PVC, steel, aluminum, brass, copper, polyethylene, boron, tantalum, shrink-tubing (teflon?), stainless steel, phenolic, neoprene, and styrofoam. The "Rack Work Summary" lists the quantities of materials used; for example, total lead was about 5.7×10^4 kg (this also appears in the Group J-6 summary), and about 454 kg of tungsten was used. The rack structure itself weighed 3.1×10^4 kg when it was "made vertical" in preparation for installing experiments, shielding, etc. The final rack weight was 1.1×10^5 kg and the harness plus cables weighed 9.8×10^4 kg. A total of 1.9×10^3 kg of Boron-Polyethylene mix was used.

c. Diagnostic Instruments

Because there is no written compilation of the materials used in the diagnostic instruments, the inventory for these materials was based on conversations with the Diagnostic Coordinator (Marion Stelts, Group P-15) and individual experimenters (Dale Glasgow - P-15 NUEX, THREX; John Stokes - P-14 HFK, Compton diodes; David Platts - CTR-1 micro-interferometry; and Michael Hynes - P-15 PINEX). As M. Hynes was on sabbatical during the inventory, contact was made with John Warren, P-15. J. Ogle of Group P-14 provided additional information concerning the amounts of fluorescing material used.

- (1) Group P-14. The Compton diode and HFK detectors used for Amarillo contained 1.0 kg of BC-400 (a polyvinyl-toluene (PVT)-based fluorescing material with a small quantity of possibly hazardous organic material).
- (2) Group P-15 - NUEX, THREX. Although gallium arsenide and indium phosphide are sometimes used, they were not employed for Amarillo. The instruments consisted of converters followed by Faraday cup or Si(Li) detectors. No hazardous materials could be identified as being associated with these detectors.
- (3) Group P-15 - PINEX. The detector used here contained 1.7 kg of BC-422 (a PVT-based fluorescing material with a small quantity of possibly hazardous organic material).

d. Composition of Device (not including Special Nuclear Materials).

This composition was based on preshot data base and drawings. Small quantities of hazardous metals (less than 1 kg) were identified in the device materials. Also, a number of organic materials were present in small quantities; except for the high explosive, none of these could be identified as hazardous. All the organic materials in the device were completely destroyed during the explosion.

Detailed device composition is classified information and is not presented here.

e. Stemming

Group J-6 provided detailed records of stemming materials. The "Stemming and Harness Diagram" shows the depths of the rack, magnetite, grout, the alternating coarse-fine layers of fill, and the series of three two-part-epoxy (TPE) plugs. "Downhole Stemming Notes" provided data on the volume of TPE and grout as well as the weight of coarse and fine fill materials. A typical truckload of grout contained 2.6×10^3 kg of grout cement, 370 kg of gel, 3.1×10^3 kg of barite, and 29 kg of D-19 (de-watering agent); seven truckloads were used. The TPE plug No. 1 used six truckloads of mix, each containing 1.3×10^3 l of epoxy mix, 5.8×10^3 kg of aggregate, and 2.3×10^3 kg of sand. Group J-6 reports the epoxy used in the TPE mix is non-hazardous .

f. Drill-back

The Fenix & Scisson Hole History indicates that the drill-back hole used 13585 bbl of bentonite and 7518 bbl of sepiolite. The hole was plugged with 70 m^3 of cement.

g. Summary of Trial Inventory

Data for all the test materials used for the selected nuclear tests at NTS were available. The data for the device, stemming, and rack were relatively easy to assimilate. Data for materials used in the diagnostic on-line detectors are less readily available. Such data can be obtained in more detail, if necessary, but with considerable effort.

Hazardous materials included the special nuclear materials in the device itself, lead, other metals, and (possibly) some organic materials used in detectors.

6. Hazardous Materials in Nuclear Tests

No attempt has been made in this review to evaluate the importance of specific hazardous material used in nuclear testing. However, it is useful to gain some perspective on this matter. The hazard caused by any material is related to its concentration when encountered—the higher the concentration, the greater the hazard. Thus, we wish to consider how materials used for testing may be diluted and dispersed during the testing process.

Subterranean test material becomes hazardous only when humans have direct contact. Thus, the presence of large amounts of lead may be considered relatively innocuous until it enters groundwater or is mined.

An underground nuclear explosion creates a volume of melted soil. A 1-kt test will melt about 1 kt of the earth's crust. Assuming an average crustal composition, about 250 kg of barium, 15 kg of gallium, and 5 kg of arsenic, for example, are naturally present in that much material. In addition, the explosion creates a chimney of fractured and crushed material with a much larger volume. This volume of fractured and crushed material is more or less directly associated with the materials added during testing: *more* from the stemming and materials at or near the top of the rack and *less* from the materials in the device and its immediate surroundings.

One perspective would be to view the amount of hazardous material added in relation to the amount naturally present in the melt or in the chimney, as a whole. From such a perspective, the use of a few kilograms or less of arsenic or gallium in a typical test can be viewed as an incremental addition to materials already existing in the geologic medium surrounding the test point.

This view is not applicable to tests with very low, or zero, energy release.

Any hazardous organic materials that are placed outside the melt zone might survive the explosion. Based on current findings, it appears that if any hazardous organic materials have been used, they have been used only in small quantities (generally less than 1-kg amounts). The importance of any such materials would be related to their actual abundance and character, neither of which have been identified.

The fissionable materials in the device and the radioactive materials produced during the explosion are clearly important hazardous materials and, with the possible exception of uranium, cannot be considered incremental to natural materials.

D. RE-ENTRY OF A CHIMNEY

There are few opportunities to sample the distribution of radionuclides in the chimney and upper cavity region at a nuclear test site. The drillbacks, which furnish the debris we described in Sec. IIIB, are done under severe time constraints and are designed to get to the shot puddle as rapidly as possible. In August 1989, Los Alamos drilling (J Division) offered to test a drill rig by drilling a hole into a collapsed zone above a cavity. The site selected was U4u, where a shot had been fired 132 m below the SWL in October 1988. Drilling operations were begun August 20; hole completion and rig demobilization were completed in 10 days. The hole U4ups2a, 25 cm in diameter, was slant drilled at about 19° from vertical. The hole measured 657 m vertical, somewhat above the puddle at the bottom of the cavity. A 9-m core was taken at a depth of 496 m; it did not contain any radioactivity. A log run inside the drill string with a NaI scintillation detector indicated regions of radioactivity from about 532 m to the bottom of the hole. (A high-intensity gamma log of the type used during drillbacks showed no radioactivity.) Eleven samples were taken with a Hunt sidewall sampling tool in the regions containing radioactivity. These samples were surveyed in the field with a germanium counter and found to contain ^{137}Cs , ^{125}Sb , and ^{106}Ru . Slots near the bottom of a 7-cm tube cemented in the hole will allow us to monitor water levels and obtain water samples. We will continue our analysis of samples.

IV. TRANSPORT

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

We described the study being carried out at the Cambric site in Sec. IIIA. Water samples collected monthly from the RNM-2S well consist of (1) a 208-l sample that is boiled to dryness and gamma counted or stored for future analysis, (2) a small-volume grab sample that is counted for tritium, and (3) a 2-l sample collected in an evacuated stainless steel tube, which is analyzed for ^{85}Kr . The samples that were boiled to dryness during the past year did not show the presence of ^{137}Cs or any other fission product; this result is the same as in previous years. The cesium ions sorb strongly on the tuffaceous alluvium, and thus, move only very slowly from the cavity region toward the pumped well. In contrast, tritium moves with the pumped water and has largely been removed from the cavity; almost 91% of the Cambric source term tritium has been pumped out of RNM-2S. Concentrations of tritium measured during the past year are given in Table XIII. The cumulative data for the elution of tritium from RNM-2S are displayed in Fig. 1. Although the concentration of tritium now being pumped from this well is relatively low, continued pumping allows us to define the tail of the elution curve and determine the hydrodynamic dispersion at this site. The pump at RNM-2S failed on September 25, 1990. It will be replaced early in FY 1991.

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

Identification Number	Date Collected	Volume Pumped (10 ⁶ m ³)	Tritium Concentration ^a (10 ⁻⁴ µCi/ml)	
			At Counting Time	At T ₀ 5/14/65
G 1497	10/17/89	15.213	4.30	17.0
G 1498	11/13/89	15.313	4.23	16.7
G 1499	12/13/89	15.407	4.15	16.6
G 1500	01/17/90	15.520	4.10	16.4
G 1501	01/26/90	15.549	4.02	16.2
G 1502	02/20/90	15.630	3.98	16.0
G 1503	03/15/90	15.704	3.82	15.7
G 1504	04/24/90	15.814	3.77	15.5
G 1505	05/24/90	15.919	3.73	15.3
G 1506	06/09/90	16.009	3.68	15.1
G 1507	07/10/90	16.076	3.64	15.1
G 1508	08/23/90	16.211	3.58	14.9
G 1509	09/25/90	16.304	3.49	14.6

^aStandard deviation of counting data < 1%.

Table XIV contains FY 1990 data reflecting analysis of the 2-*l* samples not subject to exposure to the atmosphere. In Fig. 2, we display the cumulative elution data for ⁸⁵Kr. The general shape of this elution curve is like that for tritium, although the peak occurs slightly later in time than the peak for tritium. The ⁸⁵Kr /tritium ratio has remained essentially constant over the years and is approximately one-half the value calculated for the Cambrian source term. This ratio is in contrast with that measured at RNM-1 (Ref. Sec. IIIA.), which was consistently larger than the source term value. Only about 41% of the ⁸⁵Kr initially produced by the Cambrian test has been pumped from RNM-2S. We hypothesize that krypton is transported somewhat more slowly than tritium and that an appreciable fraction of krypton is lost from solution during this transport.

B. TECHNETIUM ANALYSES (A. S. Mason and D. J. Rokop)

In our for FY 1989 annual report¹⁷, we described the analysis of several NTS water samples for ⁹⁹Tc and noted that the methodology recently developed¹⁹ at Los Alamos could be used to measure ⁹⁹Tc in RNM-2S water samples. We recently sampled RNM-2S to determine the ⁹⁹Tc content

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

Identification Number	Date Collected	Volume Pumped (10 ⁶ m ³)	Tritium Concentration (10 ⁻³ μCi/ml)	⁸⁵ Kr Concentration (10 ⁻⁷ μCi/ml)	⁸⁵ Kr/Tritium Atom Ratio ^b (x10 ⁵)	Ratio ^c
434-1-89-015	08/04/89	14.998	1.76	1.25	6.2	0.51
434-1-89-017	09/07/89	15.097	1.74	0.79	4.0	0.32
434-1-89-019	10/25/89	15.253	1.67	1.51	7.9	0.64
434-1-89-021	11/07/89	15.293	1.69	0.98	5.0	0.41
434-1-89-023	12/06/89	15.384	1.65	1.31	6.9	0.57
434-1-90-002	01/11/90	15.500	1.66	1.18	6.2	0.51
434-1-90-003	02/07/90	15.588	1.62	1.21	6.5	0.53
434-1-90-005	03/02/90	15.663	1.59	1.08	5.9	0.48
434-1-90-007	04/11/90	15.787	1.57	1.01	5.6	0.46
434-1-90-010	05/02/90	15.854	1.58	0.77	4.2	0.35
434-1-90-012	06/07/90	15.970	1.54	0.69	3.9	0.32
434-1-90-014	07/10/90	16.076	1.52	1.01	5.8	0.47
434-1-90-016	08/10/90	16.172	1.49	1.05	6.1	0.50
434-1-90-018	09/13/90	16.268	1.46	1.04	6.2	0.51

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

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

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

currently in the water. Two samples were collected from RNM-2S and one from Well 5C; the latter sample served as a blank. Each sample consisted of 1.14×10^5 l of water spiked with 100 pg of ⁹⁷Tc to serve as a tracer. The samples were passed through 50- x 15-cm-diam columns containing Purolite A-510 anion exchange resin. Because of the column flow rate, 12 hr were required to process each sample. Sampling at Well 5C went without incident, but the precipitate in the column at RNM-2S tended to obstruct the procedures. Using scanning electron microscopy, this precipitate was identified as amorphous material containing magnesium, calcium and silicon. We have not yet determined the reason it formed in our columns. The resins containing the three samples are now being processed to separate and measure ⁹⁹Tc.

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 vs volume pumped.

C. COLLOID STUDIES (E. A. Bryant)

Major personnel changes have delayed the experimental work with colloids, which we had anticipated would be started during the past fiscal year. We surveyed the literature extensively, particularly material on the collecting and characterization of colloids in environments like the NTS. During FY 1991, we will begin field work at the Cambric site using ultrafiltration to collect and concentrate colloidal materials. These samples will be returned to the laboratory for further characterization based on size, composition and charge. The interactions of the colloidal material with radioactive species and with hazardous chemicals will be evaluated. Los Alamos has considerable resources in equipment and personnel to pursue these studies. Currently within the INC division there are ongoing research projects on remediation of contaminant colloids in reservoirs and in characterization of plutonium colloids.

V. DRILLING OF UE3E#4

In Area 3 of the NTS, the HRMP is studying a site where radionuclides appear to have migrated from an identified source.^{13,14} Last year at this site, we drilled hole UE3e#4 to continue our research on the extent and mechanism of this migration. The hole, 44 cm in diameter, was drilled to 440 m; a 31-cm-diam hole was then extended to 700 m. Water was swabbed from the hole and core and sidewall samples were collected. Three piezometer tubes were placed in the hole at varying depths so that the aquifer can be monitored with respect to the hydraulic head and tritium content. Los Alamos acted as the lead organization for this drilling operation, with participation from other HRMP and NTS support organizations. We have prepared an extensive report²⁰ detailing the work at UE3e#4.

ACKNOWLEDGMENTS

We wish to express our appreciation to these individuals for their contributions to this report: M. R. Getts and R. A. Beatty for help with gamma counting; C. E. Lowe for preparing the manuscript; and M. Minahan and J. H. Heiken for editorial assistance.

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APPENDIX

HRMP Meetings Attended During FY 1990

Monterey, CA (in conjunction with Migration 89)	November 3-4, 1989
DOE-NVO, Las Vegas, NV	December 7, 1989
DOE-NVO, Las Vegas, NV	March 20, 1990
LLNL, Livermore, CA	June 25-26, 1990
DOE-NVO, Las Vegas, CA	September 5, 1990

Approximate workdays at the NTS for field sampling or drilling activities: 25 workdays

HRMP Documents Prepared During FY 1990

HRMP Budget Sheets, FY 1990 (11/13/90)

UE3e#4 Documents: Prospectus (11/17/89); Summary Report (6/19/90); Long Range Plan (7/13/90).

UE20bd Documents: 1st Draft Criteria Letter and Drilling Plan (5/90); 2nd Draft Drilling Plan (6/12/90).

“Migration of Fission Products at the Nevada Test Site: Detection with an Isotopic Tracer,” presented at Migration ‘89, Monterey, CA , November 6-9, 1989.

“Radionuclide Migration Studies at the Nevada Test Site,” presented at the International Chemical Congress of Pacific Basin Societies, Honolulu, HI, December 17-22, 1989.

The Cambic Research Experiment - A Review Paper (6/90)

Interim Report on U4ups2a (9/19/90)

Reviews prepared for NVO: Draft of HRMP Master Plan (8/1/90); Draft of Mission Statement (8/28/90)

A LIST OF HAZARDOUS MATERIALS

Organic chemicals considered hazardous and regulated under RCRA are listed under 40 CFR Parts 261 and 302; the extensive list is readily available (for example, from the Los Alamos National Laboratory Health Safety and Environment Division, HSE). Some common examples are listed below:

Acetone
Carbon tetrachloride
Chlorinated fluorocarbons
Ethyl ether
Methyl ethyl ketone
Toluene
Trichloroethylene
Xylene

Hazardous inorganic constituents include those for which the EPA has established limits for concentrations in ground waters, based upon EP-toxicity (40 CFR Chap. 1, Sec. 261.24):

Arsenic	5 mg/L
Barium	100 mg/L
Cadmium	1 mg/L
Chromium	5 mg/L
Lead	5 mg/L
Mercury	0.2 mg/L
Selenium	1 mg/L
Silver	5 mg/L

Other inorganic constituents considered hazardous include those from 40 CFR Chap. 1, Secs. 261 and 302):

Antimony
Asbestos
Beryllium
Copper
Cyanide
Fluoride
Nickel
Osmium
Radionuclides
Thallium
Zinc