*Laboratory and Field Studies Related
to Radionuclide Migration at the
Nevada Test Site*

October 1, 1997 — September 30, 1998

RECEIVED
AUG 12 1999
OSTI

Los Alamos
NATIONAL LABORATORY

*Los Alamos National Laboratory is operated by the University of California
for the United States Department of Energy under contract W-7405-ENG-36.*

Edited by Nikki Goldman, Group CIC-1

*The four most recent reports in this unclassified series are
LA-12917-PR, LA-13064-PR, LA-13270-PR, and LA-13419-PR.*

An Affirmative Action/Equal Opportunity Employer

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither The Regents of the University of California, the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by The Regents of the University of California, the United States Government, or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of The Regents of the University of California, the United States Government, or any agency thereof. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

Issued: March 1999

*Laboratory and Field Studies Related
to Radionuclide Migration at the
Nevada Test Site*

October 1, 1997 — September 30, 1998

Edited by Joseph L. Thompson

Contributors:

D. L. Finnegan

K. S. Kung

B. A. Martinez

J. L. Thompson

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

CONTENTS

EXECUTIVE SUMMARY.....	vii
ABSTRACT.....	1
I. INTRODUCTION	2
II. SAMPLE ANALYSES FROM NUCLEAR TEST SITES	2
A. VADOSE ZONE.....	2
1. Halfbeak.....	2
2. Area 3 Studies.....	3
B. SATURATED ZONE.....	6
1. Faultless.....	6
2. Shoal	6
3. Aleman	7
4. Tybo-Benham	7
C. CAVITY/CHIMNEY.....	8
1. Almendro	8
2. Dalhart.....	9
3. Cheshire	9
III. COLLOID MEASUREMENTS	13
A. PRELIMINARY STUDIES.....	14
B. ER-20-5 #1 SAMPLES	18
C. CHESHIRE SAMPLES	20
IV. PROGRAM SUPPORT ACTIVITIES	23
A. DOCUMENT REVIEW.....	23
B. SHORT COURSE ON RADIONUCLIDE MIGRATION	23
C. WORKSHOP/CONSULTATION.....	23
D. PUBLICATIONS.....	24
ACKNOWLEDGMENTS.....	24
REFERENCES	24

EXECUTIVE SUMMARY

In this report, we describe all the work done at Los Alamos during FY 1998 in support of the Hydrologic Resources Management Program and the Underground Test Area Operable Unit. Our research is done in cooperation with a number of other organizations, both federal and private, with funding from the US Department of Energy, Nevada Operations Office. This program has been in existence since 1976; its primary focus is elucidating the effects of underground nuclear testing at the Nevada Test Site (NTS). Los Alamos personnel are particularly concerned with characterizing the distribution of radioactive materials in the vicinity of expended nuclear tests and measuring the movement of such material with groundwater. Although underground nuclear testing stopped in 1992, the ongoing mission of the NTS includes environmental restoration and maintenance of test readiness. Our research program provides data that is essential for modeling radionuclide transport in groundwater at the NTS and for developing strategies for monitoring and restoration. Our personnel have the knowledge and skills required to maintain test readiness and to develop new activities at the NTS. We provide independent consultation and review for other programs at this facility, strengthening their credibility and public acceptance. The present document is the latest in our series of reports issued annually for over 20 years.

During the past year, we investigated several sites where nuclear devices had been exploded in the vadose zone above the water table. At the Halfbeak (U-19b) site, no radioactivity was found a few tens of meters below the floor of the collapse crater. The same proved to be the case in much deeper samples taken at Jerboa (U-3at-D1) and Bobac (U-3bl-D2). The lack of ^{137}Cs at Bobac was a surprise, for this site was similar in many respects to Hyrax (U-3bh), where we had found this radionuclide some five cavity radii above the working point. More drilling and sampling at these sites will be required to improve our understanding of the conditions that lead to gaseous transport of precursors to ^{137}Cs upward during chimney formation.

Water samples were collected from locations outside the immediate vicinity of several nuclear tests. At the UC-1-P-2SR well near the Faultless site in central Nevada, tritium and a low concentration of ^{137}Cs were measured. At the HC-4 well near the Shoal site in western Nevada, no tritium or other radionuclides were detected. On the NTS, we sampled water at UE3e#4 adjacent to the Aleman test. This site was studied extensively in 1985–6 because radionuclides were found before firing the Aleman shot. We determined then that the source of this material was the Sandreef event that had been fired some 350 m to the north. We continue to occasionally monitor the radionuclides at several depths at UE3e#4 but are aware that our inability to purge the residual drilling fluid at this site makes our measurements of dubious value. For several years, we have sampled water from ER-20-5 #1 and #3 wells. This location is of special interest since we have established that plutonium has migrated there not from the adjacent Tybo (U-20y) event, but from Benham (U-20c) about 1.3 km to the north. This year, we again confirmed our previous measurements and added ^{241}Am to the list of radionuclides detected in water from ER-20-5 #1. Its concentration was quite low, several orders of magnitude less than that of the plutonium. This radionuclide has rarely been measured in groundwater at the NTS.

In spite of our long history of water sampling at the NTS, we have had relatively few opportunities to collect water directly from the cavity or chimney formed by a nuclear test. This year, we sampled water from three holes penetrating the cavity or chimney at nuclear test sites. At the Almendro (U-19v) site, we measured tritium and ^{85}Kr in the water, which remains at a high temperature in the cavity region. At Dalhart (U-4u ps 2a), we used for the first time tandem Bennett pumps to extract water from the drill back hole that extends into the upper chimney. Pumping was quite slow, and the hole was not completely purged before the sample was taken. We expect to pump for a longer period and collect samples more representative of formation water at the sampling depth. The application of tandem Bennett pumps to purge small diameter tubes is a very important advance for our program, for there are a number of wells at the NTS that were completed by installing 7.3-cm OD piezometer tubes for access to the groundwater. At the Cheshire (U-20n ps1 ddh) well, pump speed was not an issue. This well is one of the most studied sites on the NTS and one that continues to yield information vital to our program. The Cheshire well had been configured to sample water only from a horizon several hundred meters above the working point and had not been pumped since 1985. This year, we pumped more than enough water from the well to purge it, then collected samples representative of the upper horizon. Next, the well was reconfigured with a straddle packer so that water was drawn only from the horizon near the working point (about 1167 m vertical depth). After nearly 10^3 m^3 of water were pumped out, we collected samples representative of the cavity water. The analyses of these samples are now in progress, and we are very interested to learn what radionuclides are present in water that has been in contact with cavity melt glass for over twenty years.

This year, we put a considerable effort into analyzing the colloid content of groundwater from the NTS. This work required us to evaluate possible sources of colloid contamination associated with collection, filtering, analyzing, and storing of water samples. We evaluated the colloid concentrations and size distributions in water samples from the ER-20-5 and U-20n ps1 ddh wells using a particle measuring spectrometer, which detected colloids in the size range 50 to 200 nm (as well as $>200 \text{ nm}$). Both these sites produced groundwater with an abundance of colloids in the detection range. The two zones sampled at ER-20-5 were rather similar in colloid content, but at U-20n ps1 ddh, the deeper zone had 20 times the colloid content of the upper zone. We observed that filters with relatively large pore sizes (200 and 450 nm) trapped many colloids of dimensions as small as our instrument could detect. This result was especially true if the filter was excessively loaded with particulates. Electron microscopy showed that our filtered colloids came in a wide variety of sizes and shapes. These studies underscore the difficulty we have in determining the relationship between the radioactivity and the colloid content of our NTS groundwater samples.

We reviewed this year a number of documents produced for the NTS Environmental Restoration program. Most of these were related to efforts to model radionuclide transport. Our concern is that there is an acute shortage of data about the concentrations of a number of radioactive species possibly in groundwater in test cavities. Until we know more about the hydrologic source term, it is hard to model transport with any confidence. Los Alamos personnel participated in the second short course on radionuclide transport given at Las Vegas, this time videotaping the presentations so that they may be used for future educational efforts. We also produced several publications on our work. One of these is in a prestigious international journal and should bring attention to the NTS as a place offering unique opportunities to study contaminant transport in groundwater.

LABORATORY AND FIELD STUDIES RELATED TO RADIONUCLIDE MIGRATION AT THE NEVADA TEST SITE

October 1, 1997 – September 30, 1998

Joseph L. Thompson, Editor

ABSTRACT

In this report, we describe the work done in FY 1998 at Los Alamos National Laboratory as part of the Hydrologic Resources Management Program (HRMA) funded by the Nevada Operations Office of the US Department of Energy (DOE/NV). The major part of our research effort was to measure radionuclides present in water or soil samples collected from near nuclear tests. We report our measurements for materials collected in both saturated and unsaturated horizons adjacent to nuclear test cavities or collapse chimneys and from within several cavities. Soil samples collected from above the cavities formed by the Halfbeak, Jerboa, and Bobac tests contained no radioactivity, although a test similar to Bobac in the same area had been contaminated with ^{137}Cs . Water samples from near the Shoal test contained no measurable radionuclides, whereas those from near Faultless and Aleman had concentrations similar to previous measurements. Water from the Tybo-Benham site was similar to earlier collections at that site; this year, we added ^{241}Am to the list of radionuclides measured at this location. Two Bennett pumps in tandem were used to extract water from the piezometer tube in the cavity of the Dalhart event. This extraction is a significant achievement in that it opens the possibility of purging similar tubes at other locations on the NTS. The Cheshire post shot hole was reconfigured and pumped from two horizons for the first time since mid-1980. We are especially interested in examining water from the level of the working point to determine the hydrologic source term in a cavity filled with groundwater for over 20 years. We devoted much time this year to examining the colloid content of NTS groundwater. After developing protocols for collecting, handling, and storing groundwater samples without altering their colloid content, we analyzed water from the Tybo-Benham and from the Cheshire sites. Whereas the colloid concentration did not vary much with depth at Tybo-Benham, there were 20 times more colloids in groundwater from the Cheshire cavity than were found a few hundred meters higher. Electron micrographs show the wide variety of colloid sizes and shapes present in NTS groundwater. Our experiences with filtration of groundwater samples illustrate the difficulties of colloid size characterization using this methodology. Our report ends with a description of our consultative and educational activities and a list of recent publications.

I. INTRODUCTION

Los Alamos National Laboratory is one of several organizations participating in the Hydrologic Resources Management Program (HRMP) and the Underground Test Area Operable Unit (UGTA) sponsored by the US Department of Energy, Nevada Operations Office (DOE/NV). This report describes the work done at Los Alamos in support of the HRMP and UGTA during FY 1998. After outlining the role of the Laboratory in this work and indicating our relationships to other participating organizations, we report the results of analytical work on solid and liquid samples collected at the Nevada Test Site (NTS). Also, we describe program support activities to which we contributed during this fiscal year and list publications resulting from our work.

In 1973, Los Alamos National Laboratory (LANL) and Lawrence Livermore National Laboratory (LLNL) began collaborating on a study of the geological and hydrological consequences of underground testing of nuclear devices at the NTS. We sought to discover whether radionuclides associated with this testing were mobile in groundwater and could be moved appreciable distances from detonation sites. These studies are still going on; the laboratories now work with the US Geological Survey (USGS), the Desert Research Institute (DRI), and several NTS contractors such as Bechtel Nevada Corporation (BN) and International Technology Corporation (IT). We have studied a number of test locations in great detail; several locations have been sampled for more than twenty years. Although underground testing at the NTS ended in 1992, the need for our work continues. Our data information base and the personnel expertise developed over nearly three decades are useful to several NTS programs, especially Defense Programs, and Environmental Restoration. Stockpile stewardship, maintenance of test readiness, and protection of land and water resources are issues important to Defense Programs, and HRMP personnel are able to contribute to NTS work on these subjects. The Environmental Restoration program (UGTA) is concerned with modeling radionuclide transport on and off the NTS, with monitoring and sampling waterborne radioactive material, and with understanding the effect of the NTS on regional groundwater—all topics on which we have some expertise. Our reports produced over the past 25 years are an indication to the public that the DOE/NV takes seriously its responsibility to manage the water and land resources of the NTS.

This report is the latest in a series of annual reports.¹⁻²¹ We have also produced a number of topical reports²²⁻²⁷ and journal articles.²⁸⁻³⁹

II. SAMPLE ANALYSES FROM NUCLEAR TEST SITES

A. Vadose Zone

1. Halfbeak (U-19b) This shot was fired at a depth of 820 m on 30 June 1966 in Area 19 at Pahute Mesa. The standing water level at this site was 646 m. In mid-1997, personnel from DRI auger drilled into the collapsed crater of Halfbeak as part of a study of moisture infiltration. Samples of rock were collected at 1.5-m intervals to a depth of

24.4 m and saved for a radiochemical survey of gamma emissions. In December 1997, we received 16 boxes (~ 9.5 cm × 6 cm × 3 cm) containing these samples. The rock was unconsolidated; pieces ranged in size from that of marbles to coarse dust. We placed each box on a Ge detector and counted it for 100 minutes. The gamma spectra showed no indication of any radioactivity other than that associated with naturally occurring uranium, thorium, and potassium. We estimate that we could detect as little as 37 Bq/kg (1 pCi/g) of ^{137}Cs in these samples. These data suggest that there is no radioactivity associated with the Halfbeak event at shallow depths below the collapse crater.

2. Area 3 Studies In 1996, LANL and LLNL cooperated in analyzing cores derived from a subsidence crater in Area 3 of the NTS. The crater, which had resulted from the 1962 Hyrax test at U-3bh, was being studied for possible use in low-level radioactive waste disposal. The results of core analyses have been published in several reports.^{27,40} A significant observation was that ^{137}Cs had moved upward at this site to a horizon some 5 cavity radii above the Hyrax working point. We interpreted this movement to have been the result of gaseous transport of volatile precursors of ^{137}Cs (accompanied by CO_2), which occurred as the chimney collapsed about eight minutes after cavity formation. In 1998, LANL personnel obtained cores from two other subsidence craters in Area 3. These craters had been formed from tests similar to Hyrax in that their working points were above the water table, in alluvium, and the devices had been emplaced with similar methodology. One of these tests, Jerboa, collapsed 35 minutes post shot. The other, Bobac, collapsed 8 minutes post shot. We anticipated that the upward migration of ^{137}Cs at Bobac would be similar to that at Hyrax and that this migration would not have taken place at Jerboa because of the longer time before the crater collapsed. Here we report our analyses of cores from Jerboa and Bobac and discuss the significance of our findings.

Jerboa (U-3at-D1) The Jerboa test was conducted 1 March 1963 at a depth of 310 m at U-3at. Cavity collapse occurred 35.3 minutes post shot; the collapse zone extended to the surface and formed a crater about 32 m deep. No release of radioactivity was detected. In June 1996, slant holes were drilled under the collapse crater at U-3at and core samples obtained. The longer of these slant holes (U-3at-D1) extended to a vertical depth of 147 m. Our report concerns six cores from this hole collected from vertical depths of 56 to 147 m. Drilling and coring were done with an air percussion hammer, so no fluids were introduced. A complete description of the drilling and coring operation is given in Reference 41. The cores were contained in 89-mm OD polycarbonate tubes 76 mm long. These tubes were fitted with end caps, taped, and sealed in a plastic laminate. We received these cores in February 1998, so they had been in storage about 16 months. We determined the amounts of tritium and gamma-emitting radionuclides in each core. After the core was removed from its container, a portion was heated in an evacuated system and the extracted water collected in an ice trap. Aliquots of this water were analyzed for tritium using a liquid scintillation counter. Portions of the dried core were analyzed with a germanium detector system to determine the quantity of gamma-emitting radionuclides present. Our tritium results are shown in Table I below. Very small amounts of tritium were detected in the three deeper cores. Our data are in agreement with the tritium analyses done by DRI and reported by BN in Reference 41; their methodology employed tritium enrichment, which allowed lower concentrations to be measured. We found amounts of water in

the cores similar to those reported by BN (their data are in volume percent while ours are in weight percent). These data indicate that the cores did not lose significant amounts of water during their period of storage. We detected no gamma activity other than that from naturally occurring radionuclides. Our sensitivity for ^{137}Cs in samples of this size is on the order of 2E-3 Bq/g .

Table I. Water and tritium in cores from U-3at-D1. Comparison data from Reference 41 was converted to Bq/L at t_0 for the Jerboa test.

Sample ID	Vertical Depth (m)	% Water (by weight)	^3H (pCi/ml) at count time	^3H (Bq/L @ t_0)	
				LANL	Bechtel
332-96-310	56	5.3	not detected	not detected	$0.3 \pm 300\%$
332-96-320	87	4.2	not detected	not detected	not detected
332-96-330	126	8.3	not detected	not detected	$11 \pm 100\%$
332-96-340	131	8.5	$9\text{E-1} \pm 20\%$	$2\text{E2} \pm 20\%$	$1\text{E2} \pm 84\%$
332-96-350	136	10.4	$9\text{E-1} \pm 33\%$	$2\text{E2} \pm 33\%$	$2\text{E2} \pm 32\%$
332-96-360	147	6.0	$1.9\text{E0} \pm 18\%$	$5\text{E2} \pm 18\%$	$6\text{E2} \pm 13\%$

Bobac (U-3bl-D2) The Bobac test was conducted 24 August 1962 at a depth of 195 m at U-3bl. Cavity collapse occurred 8.2 minutes post shot; the collapse zone extended to the surface and formed a crater about 14 m deep. No release of radioactivity was detected. The collapse crater was immediately adjacent to a slightly larger and deeper crater formed from the Paca test conducted 7 May 1962 in U-3ax. The crater at U-3ax was used for disposal of contaminated soil and equipment starting in the late 1960's; similar use of U-3bl started in 1984. Very little liquid was disposed of in these craters and a covering of alluvial soil was emplaced in 1987. Before the present study, excavation of material between the two collapse craters formed a single unit with a volume in excess of 2E5 m^3 . More details about this area are given in Reference 42. In February 1996, a slant hole was drilled from the edge of the U-3bl crater at about 45° so as to pass under the collapse zone and through the rubble chimney below it. This borehole (U-3bl-D2) extended across the estimated boundaries of the chimney and terminated at a vertical depth of approximately 97 m. Twenty-nine cores were collected at 3-meter intervals through the chimney region. The drilling and coring were done with an air percussion hammer with no introduction of fluids. A more complete description of the drilling and coring operation is given in Reference 42. Cores were contained in 89-mm OD polycarbonate tubes 76 mm long; the tubes were fitted with end caps, taped, and sealed in a plastic laminate.

In February 1998, we selected ten cores from the slant hole through the U-3bl chimney for analysis; they had been in storage for two years. We measured the tritium and gamma-emitting fission products in these cores. The material from the core container was heated in an evacuated system and the extracted water collected in an ice trap. Aliquots of this water were analyzed for tritium using a liquid scintillation counter. Portions of the dried core were analyzed with a germanium detector system to

determine the quantity of gamma-emitting radionuclides present. The tritium analyses are shown in Table II below. The values are quite low but above background levels. They are in general agreement with the analyses reported by BN (Reference 41); these analyses had been done by DRI using tritium enrichment. We also found amounts of water similar to those reported by BN, though our water is reported as weight percent rather than volume percent. This result implies that the cores did not lose a significant amount of water during the two-year storage. We could detect no gamma activity other than that attributable to naturally occurring radionuclides. Sensitivity to gamma-emitters is rather high; for example, we can detect ^{137}Cs in amounts of 2E-3 Bq/g .

Table II. Water and tritium in cores from U-3bl-D2. Comparison data from Reference 42 was converted to Bq/L at t_0 for the Bobac test.

Sample ID	Vertical Depth (m)	% Water (by weight)	^3H (pCi/ml) at count time	^3H (Bq/L @ t_0)	
				LANL	Bechtel
296-96-300	44	6.8	3.3	$8.8\text{E}2 \pm 10\%$	$1.2\text{E}3 \pm 8\%$
296-96-310	50	6.2	2.3	$6.3\text{E}2 \pm 10\%$	$8.1\text{E}2 \pm 8\%$
296-96-320	56	8.7	2.0	$5.6\text{E}2 \pm 8\%$	$7.1\text{E}2 \pm 10\%$
296-96-330	62	8.0	2.4	$6.5\text{E}2 \pm 7\%$	$9.2\text{E}2 \pm 8\%$
296-96-340	69	5.5	3.7	$1.0\text{E}3 \pm 7\%$	$1.5\text{E}3 \pm 4\%$
296-96-350	75	9.6	11.3	$3.1\text{E}3 \pm 5\%$	$3.0\text{E}3 \pm 3\%$
296-96-360	81	7.4	4.6	$1.3\text{E}3 \pm 8\%$	$1.5\text{E}3 \pm 7\%$
296-96-370	87	5.8	3.2	$8.6\text{E}2 \pm 10\%$	$1.1\text{E}3 \pm 5\%$
296-96-380	93	9.8	5.1	$1.4\text{E}3 \pm 6\%$	$1.6\text{E}3 \pm 6\%$
296-96-390	97	10.4	4.6	$1.3\text{E}3 \pm 8\%$	$1.4\text{E}3 \pm 7\%$

The study of core material from Jerboa and Bobac was undertaken to learn if ^{137}Cs was distributed upward from the cavity as was the case at Hyrax. All three tests had been conducted in sandy alluvium; the Jerboa test had a working point about 100 m deeper than the other two and its cavity did not collapse until the precursors to ^{137}Cs had decayed to this nongaseous radionuclide. The collapse times for Hyrax and Bobac were approximately eight minutes. At each test, the device was covered with magnetite sand, then with alternating layers of coarse and fine sand to the top of the emplacement hole. Based on this information, we expected that core samples collected from about five cavity radii above the working point would contain ^{137}Cs in the case of Bobac and not in the case of Jerboa. Device-related tritium or fission products other than ^{137}Cs were not expected.

At Jerboa, we detected no ^{137}Cs in any cores. There was a slight increase in the tritium over ambient levels in the very deepest samples (at about 4 cavity radii above the working point). The absence of ^{137}Cs was expected; the hint of tritium from the device was not. In general, we have not observed tritium at distances more than about 2.5 cavity radii from cavities unless it moved with flowing groundwater. But these tests provide our first chances to study tritium distribution in the vadose zone in alluvium,

so we have no data for comparison. It would be very interesting to drill closer to the cavity at Jerboa to better define the deposition of tritium around the cavity boundary.

At Bobac, we detected no ^{137}Cs even in cores taken about 3.5 to 4.5 cavity radii above the working point. The tritium content was quite low in all the cores. We are left with a puzzle as to why the ^{137}Cs distribution is different from that at Hyrax when the two tests appeared to be so similar. Drilling deeper into the chimney at Bobac and also at Hyrax would provide more data to help us understand the distribution of both tritium and ^{137}Cs around the cavities and chimneys of tests conducted in the vadose zone. The present limited study reveals how little we know about this subject.

B. SATURATED ZONE

1. Faultless (UC-1-P-2SR) The Faultless test was conducted in central Nevada on 19 January 1968. On 23–24 October 1997, personnel from DRI and LLNL collected water samples from the drill back hole at this site, UC-1-P-2SR. Several of these samples were shared with Los Alamos; we report in Table III below the results of our analyses of them.

Table III. Tritium activity in Faultless water samples. Activities are corrected to $t_0 = 19 \text{ Jan } 1968$.

Sample ID	Description	Tritium Activity (Bq/L)
1968-97-111	795 m, trip #1, 23 Oct	2.91E6
1968-97-112	795 m, trip #2, 23 Oct	2.85E6
1968-97-113	795 m, trip #3, 24 Oct	2.91E6

A composite (1968-97-110) was made of several samples of water collected from the drill hole at a measured depth of 795 m (2607 ft); this water (9.85 L) was evaporated and the residue counted for 3000 min on an intrinsic germanium detector. The only gamma-emitting, nonnatural radionuclide identified was ^{137}Cs . Its concentration was 0.10 Bq/L at count time, or 0.2 Bq/L at t_0 .

2. Shoal (HC-4) The Shoal test was fired 26 October 1963 near Fallon, NV, with an announced yield of 12 kt. One of the wells in the vicinity of this test is HC-4. On 16 January 1998, personnel from DRI and LLNL sampled water from this well after purging the well at 1 gal/min for two days. A 208-L barrel of water was sent to Los Alamos; it arrived 21 January 1998, and analysis was begun a few days later (assigned ID 1969-98-110). We took aliquots for tritium measurement by liquid scintillation counting and evaporated the bulk of the sample (207 kg) to dryness. An aliquot of the dried residue was counted on a calibrated germanium detector system to quantify any gamma-emitting radionuclides. No tritium was detected above background levels. Our detection level is about $3.7\text{E}1 \text{ Bq/L}$. A previous analysis by DRI of a water sample collected from this well in February 1997 was $4.2\text{E}1 \text{ Bq/L}$. Presumably, this analysis was done using an enrichment procedure, which gives better sensitivity than our method. Our data suggest that the tritium concentration has not appreciably increased since 1997 in well HC-4 but do not allow us to judge if it has decreased. No gamma-

emitting radionuclides other than natural background constituents were detected. Our sensitivity for ^{137}Cs is about $3.7\text{E-}4$ Bq/L in a sample of this type.

3. Aleman UE3e#4 In 1985 and 1986, the emplacement hole U-3kz was being prepared for the nuclear test Aleman, and radionuclides were found in water pumped from the hole. Los Alamos personnel were able to establish that the origin of this radioactivity was the test Sandreef (U-7aq) about 350 m to the north.^{26,31} In the course of investigating the Aleman site, several exploratory holes were drilled. UE3e#4 was the fourth of these.²³ It was completed by cementing three slotted piezometer tubes in the hole with openings at the indicated depths: tube 1, 655–662 m; tube 2, 575–582 m; tube 3, 493–500 m. Water pressures increase with depth in this area so that the standing water levels in the deepest piezometer tube is higher than in the shallowest piezometer tube. This site was last sampled in 1993.¹⁸ On 23 and 24 September 1998, personnel from LANL, LLNL, and USGS sampled water from UE3e#4 using 2-L pressure tubes. As we found in previous visits to this site, the water in the piezometer tubes had a high particulate content. In fact, the two pressure tubes used to sample piezometer tube 3 became plugged with clay-like material, and only 100 mL of water were collected. This water was given to LLNL for their analyses. The results of our analyses are given in Table IV. Our experience this year at this site confirms the necessity to pump out residual fluids from piezometer tubes before collecting water samples. We continue to experiment with Bennett pumps in tandem as a means of accomplishing this task.

Table IV. Water samples from UE3e#4. Concentrations of ^3H and ^{85}Kr are corrected to the Sandreef t_0 (9 Nov 1977).

Sample ID	Piezometer Tube #	Sample Depth (m)	Water Level (m)	^3H (Bq/L)	^{85}Kr (Bq/L)
8881-98-210	1	658	330	9.29E5	sample lost in processing
8882-98-210	2	579	404	1.9E3	2.2E2
8883-98-210	3	500	473	no sample	no sample

4. Tybo-Benham (ER-20-5 #1, #3) The ER-20-5 wells are located about 280 m southwest of the Tybo test (U-20y) and 1300 m south of the Benham test (U-20c). Water samples collected from the #1 and #3 wells contained radionuclides such as tritium, ^{60}Co , ^{137}Cs , and several europium isotopes that may be expected to be found close to nuclear test cavities. However, these water samples also contained low concentrations of plutonium with an isotopic “fingerprint” that enabled us to determine that the plutonium came from the Benham test (and not from Tybo or other adjacent tests). We found that the plutonium was associated with filterable colloids at the ER-20-5 sampling site, a discovery suggesting that at least part of its movement from Benham may have involved colloid transport. Details of our studies at this location are given in our FY 1997 annual report²¹ and in a journal article.³⁹ We continue to study this site as it provides a unique opportunity to observe the movement of plutonium with groundwater for distances over a kilometer. Both the #1 and #3 wells were pumped and sampled in 1998. The radionuclide concentrations from #1, as shown in Table V,

were similar to those reported previously.²¹ The t_0 tritium concentration in #3 after pumping $1.70 \text{ E}2 \text{ m}^3$ was $1.87\text{E}4 \text{ Bq/L}$, very close to previous values.²¹

We continued the analysis of the water sample collected from ER-20-5 #1 on 22 April 1997 and determined the concentration of ^{241}Am . It was $2.4\text{E-}14 \text{ g/L}$ and $3.1\text{E-}3 \text{ Bq/L}$ ($8.4\text{E-}2 \text{ pCi/L}$). The small mass of ^{241}Am —a factor of 300 less than that of the plutonium in the same sample—made it difficult to measure. This test is one of the few times that this radionuclide has been unambiguously measured in water samples from the NTS.

Table V. Tritium and gamma emitters in ER-20-5 #1 water (unfiltered).

Radionuclide	Activity at count time		Activity at $t_0 =$ 14 May 75		% Error
	Bq/L	pCi/L	Bq/L	pCi/L	
<i>9 Jul 98 after pumping of $1.10 \text{ E}2 \text{ m}^3$</i>					
^3H	2.33E6	6.3E7	8.6E6	2.33E8	± 2
^{137}Cs	4.88E-1	1.32E1	8.33E-1	2.25E1	± 1
^{60}Co	4.7E-1	1.3E0	1.0E0	2.7E1	± 5
^{152}Eu	4.8E-2	1.3E0	1.6E-1	4.3E0	± 5
^{154}Eu	5.4E-2	1.4E0	3.5E-1	9.5E0	± 8
^{155}Eu	1.1E-2	2.9E-1	3.2E-1	8.6E0	± 15

C. CAVITY/CHIMNEY

1. Almendro (U-19v) The Almendro test took place 6 June 1973. The re-entry hole is cased with 168-mm pipe extending to near the bottom of the cavity, which is at about $1.16\text{E}3 \text{ m}$ vertical. Although the cavity/chimney is filled with water, it has remained quite hot. The temperature of the water was so high that we could not sample it until 1993 and then not to the bottom of the hole. On 16 December 1996, Atlas Wireline Services ran a probe to about $1.09\text{E}3 \text{ m}$ vertical and measured the temperature to be 157°C . On 22 September 1998, personnel from LANL, LLNL, and the USGS collected water samples in 2-L pressure tubes at a slant depth of $1.09\text{E}3 \text{ m}$ ($1.02\text{E}3 \text{ m}$ vertical). The water level was about $6.19\text{E}2 \text{ m}$ vertical. Our analyses of these samples are shown in Table VI. The tritium concentrations are almost identical to those measured in the 1996 sampling at similar depths.²⁰

Table VI. Tritium and ^{85}Kr activities in water from U-19v. Activities are corrected to the Alameda t_0 (6 June 1973).

Sample ID	Sample Date	Sample Depth Vertical (m)*	^3H (Bq/L)	^{85}Kr (Bq/L)
7910-98-210	22 Sep 98	1.02E3	2.89E7	1.0E4
7910-98-220	22 Sep 98	1.02E3	2.87E7	sample lost in processing

*This assumes slant depth $\times 0.936$ = vertical depth.

2. Dalhart (U-4u ps 2a) The Dalhart test was conducted in 1988 at a vertical depth of 640 m, well below the preshot standing water level of 508 m. In 1990, U-4u ps 2a was drilled in the chimney region and a 7.3-cm OD tube inserted with slots from 472 to 501 m vertical. Water samples collected in 2-L pressure tubes from this site in 1992 and 1993 contained high concentrations of particulates.¹⁸ In 1995, we tried unsuccessfully to clean the hole by air-jetting fluid out.¹⁹ Water samples collected in pressure tubes in 1997 still contained much particulate matter.²¹ This hole was a candidate for purging using two Bennett pumps in tandem, as described in our FY 1997 report.²¹ On 23 September 1998, personnel from BN collected a 208-L drum of water from U-4u ps 2a after pumping a few cubic meters of water from the hole. We anticipate collecting more samples after further pumping to more completely purge residual drilling fluids from the hole. Such an operation will require considerably more pumping time since the Bennett pumps deliver about 2 liters per minute. The results of the September sampling are given in Table VII. We have included in this table values from 1997 (the ^{85}Kr has not been reported previously). So far we have detected only relatively mobile radionuclides at this site, as we would expect in samples collected in the chimney region.

Table VII. Radionuclides in water from U-4u ps 2a. All activities are corrected to t_0 = 13 October 1988.

Sample ID	Sample Date	^3H (Bq/L)	^{85}Kr (Bq/L)	^{60}Co (Bq/L)	^{125}Sb (Bq/L)	^{137}Cs (Bq/L)
1112-97-211	21 Jul 97	9.7E5				
1112-97-210	21 Jul 97		3.2E2			
1112-97-221	21 Jul 97	9.9E5				
1112-97-220	21 Jul 97		4.6 E2			
1112-98-120	23 Sep 98	1.40E6		1.0E0	3.33E1	3.26E0

3. Cheshire (U-20n) The Cheshire site was studied in 1976 and in 1983–7 to measure the migration of radionuclides from the cavity/chimney region. We utilized a drill back hole (U-20n ps1 ddh) to collect water samples. During 1983–4, a total of 1.3E4

m³ of water was pumped from perforations in the liner of this slant hole that passed through the cavity near the working point (at 1167 m vertical depth or about 1230 m slant depth). In 1985, a bridge plug was set above these perforations, and new perforations were made over the interval 812 to 913 m slant depth, corresponding to a zone of high hydrologic transmissivity in the surrounding rock strata. A total of 2.2E4 m³ of water was pumped from this upper horizon. In 1987, a down gradient hole (UE-20n #1) was drilled about 300 m southwest of U-20n ps1 ddh, and 1.7E3 m³ of water were pumped from it. Water samples were collected during all of these pumping campaigns, and a number of radionuclides were identified. Filtration experiments established a linkage between colloids and radionuclide migration. Detailed reports concerning our work at Cheshire are given in references from LANL⁷⁻¹¹ and LLNL.^{43,44} Also, a summary report on our work at this site has been written and is in review.⁴⁵ Pumping operations ceased at the Cheshire site when the down gradient hole became contaminated with ferric hydroxide, but we continued to sample water in U-20n ps1 ddh with bailers (2-L stainless steel pressure tubes) on an occasional basis. Our studies at the Cheshire site gave us insights concerning radionuclide movement away from large, below-the-water-table nuclear tests situated in the fractured rhyolite and layered tuffs of Pahute Mesa. In 1998, we revisited this site to determine how the concentrations of the radionuclides in the hydrologic source term had changed and in the hope that we could better characterize actinide speciation in collected samples.

Chimney samples In July 1998, a pump was inserted in U-20n ps1 ddh, and about 3.9E1 m³ of water were pumped out; samples were collected in 208-L drums and in 2-L pressure tubes. This water was drawn in through the existing perforations over the interval 812 to 913 m slant depth; radionuclide concentrations are comparable with those measured in the same upper zone of transmissivity since mid-1985. Tritium was measured from both the drum and pressure-tube samples; ⁸⁵Kr was separated and quantified from the pressure-tube samples. The gamma-emitting radionuclides were measured in the dried residue from the drum samples. In Tables VIII and IX, our data from the July 1998 sampling are compared with similar results from previous measurements. All data are referenced to the Cheshire shot time 14 February 1976.

Table VIII. Representative ³H and ⁸⁵Kr concentrations in water from the interval 812 to 913 m slant depth in U-20n ps1 ddh (referenced to t₀ = 14 February 1976).

Sample Date	³ H (Bq/L)	⁸⁵ Kr (Bq/L)
1985 (pumped)	1.52E7	9.6E3
1988 (bailed)	7.8E6	7.8E2
1994 (bailed)	6.6E6	(4-7)E1
1996 (bailed)	(3-7)E6	(0.1-5)E3
1997 (bailed)	(3-6)E6	1.6E1
1998 (pumped)	(4-5)E6	2.5E3

The data in Table VIII show that the tritium concentration dropped by a factor of two after pumping was stopped in 1985 and then remained essentially constant for a decade. Even when pumping was resumed in 1998, the concentration was little

changed from recent bailed samples. The ^{85}Kr concentrations were much more variable; successive bailed samples gave widely differing results depending in part on the depth from which the sample was withdrawn and whether the water in the pipe had been disturbed by previous bailer insertions. However, comparison of the *pumped* samples shows that the ratio of tritium to krypton is very similar in 1998 to what it was in 1985. We believe these data illustrate the difficulties of obtaining representative samples with bailers in small-diameter holes; they also indicate that ^{85}Kr and tritium are equally conservative water tracers over a decade.

Table IX. Representative gamma-emitting radionuclides in water from the interval 812 to 913 m slant depth in U-20n ps1 ddh (referenced to $t_0 = 14$ February 1976).

Sample Date	^{60}Co (Bq/L)	^{125}Sb (Bq/L)	^{137}Cs (Bq/L)
1985 (pumped)	5E-3	1.1E2	2.0E1
1988 (bailed)	not detected	7E0	4E-1
1994 (bailed)	3E-1	2E1	6.7E-1
1996 (bailed)	trace	2E1	6.7E-1
1997 (bailed)	not detected	not detected	6.7E-1
1998 (pumped) #8521-98-150	not detected	5.5E0	2.6E-1

The data in Table IX indicate that the two gamma-emitting radionuclides frequently found in the upper regions of the U-20n ps1 ddh borehole are ^{125}Sb and ^{137}Cs . Also, ^{60}Co appears to be present sometimes at very low concentrations. Europium isotopes ($^{152,154,155}\text{Eu}$) were found in quite low concentrations in 1985 during pumping⁹ but not since. Comparisons of the levels of ^{125}Sb and ^{137}Cs in *pumped* samples show that both these radionuclides have considerably diminished in concentration from 1985 to 1998, the antimony by a factor of 20 and the cesium by a factor of 85. The antimony is believed to be present as an antimonate anion that does not sorb readily on the rock surfaces, whereas cesium does sorb strongly on NTS rocks. We note that the conservative radionuclides tritium and ^{85}Kr decreased in concentration by a factor of less than 4 over the same period.

In 1985, plutonium was reported⁹ to be in chimney water, but it was at such low concentrations (4-7E-3 Bq/L) that we were not sure it was really present. Since that time, our analytical sensitivity for this element has improved, and we report with confidence that plutonium is present in water from this horizon; our values for plutonium are 1.4E-4 Bq/L ($\pm 15\%$).

Cavity samples In August and September of 1998, the liner in U-20n ps1 ddh was configured to allow water samples to be withdrawn from the cavity horizon. The plug at 945 m slant depth was milled out; the liner was perforated over the interval 1244 to 1253 m slant depth; and a bridge packer was set from 789 to 931 m slant depth. These changes made it possible to pump water in through the bottom perforations without a contribution of water from the perforations higher in the liner. A pump with an intake at 767 m slant depth and capacity of about 30 gpm began operation in mid-September.

About 8.12E2 m³ of water were pumped before sample collection. We believe this was enough to flush out the lower horizon of the well and to offset any effect of the 5.3E2 m³ of water added to the top of the well during the milling operation. Samples were collected in 2-L pressure tubes and in 208-L drums; also, grab samples were taken in 1-L bottles for colloid analyses. The 2-L samples were analyzed for tritium and ⁸⁵Kr. The 208-L samples were taken to dryness and the residue analyzed for gamma-emitting radionuclides. This residue was then further analyzed by alpha spectroscopy and mass spectrometry for actinides. In Tables X and XI, we show our data from the 1998 samples in comparison with values from the last samples taken at this depth in 1984.

Table X. Representative ³H and ⁸⁵Kr concentrations in water pumped from the interval 1244 to 1253 m slant depth in U-20n ps1 ddh (referenced to t₀ = 14 February 1976).

Sample ID	³ H (Bq/L)	⁸⁵ Kr (Bq/L)
1984 (852-9-84-003)	1.92E7	1.0E4
1998 (8521-98-180)	9.36E6	sample lost in processing
1998 (8521-98-251)	9.29E6	sample lost in processing

Our data indicate that the tritium concentration at the cavity horizon has dropped by about a factor of two during the interval 1984 to 1998. Unfortunately, the krypton samples from 1998 were lost in processing, so we do not know if there is a corresponding decrease in the concentration of ⁸⁵Kr.

Table XI. Representative concentrations of gamma-emitting radionuclides in water pumped from the interval 1244 to 1253 m slant depth in U-20n ps1 ddh (referenced to t₀ = 14 February 1976).

Sample ID	⁶⁰ Co Bq/L	¹²⁵ Sb Bq/L	¹³⁷ Cs Bq/L	¹⁵² Eu Bq/L	¹⁵⁴ Eu Bq/L
1984 (852-9-84-003)	5.2E-3	1.0E2	5.6E1	4E-3	2E-2
1998 (8521-98-180)	2.2E-1	1.3E2	9.6E1	1.4E-1	1.6E-1

The gamma-emitting radionuclides appear to have increased in concentration during the 14-year interval, in contrast with the decreased concentration seen for tritium. The increase is rather small for ¹²⁵Sb and ¹³⁷Cs, but large for the europium isotopes and ⁶⁰Co. However, the 1984 values of the europium concentrations were order of magnitude only, so we do not really know the extent of their changes. Perhaps the concentrations of ¹²⁵Sb and ¹³⁷Cs are increasing because of dissolution and desorption of materials in the cavity region. We hope to gain more information about

the speciation of these radionuclides by filtration studies on duplicate samples. Actinide analyses (plutonium and americium) for these samples are in progress.

III. COLLOID MEASUREMENTS

(Editor's note: The work reported in this section was done by K. S. Kung, who was studying colloids in water systems of interest to the DOE/NV Yucca Mountain project. Our program was fortunate to be able to benefit from the presence of this related research effort.)

Studies of water samples provided by the HRMP sought to (1) quantify the total colloid concentration, (2) evaluate the colloid size distribution, and (3) determine colloid shapes and mineral compositions. The colloidal particle size distribution and total particle concentration between 50- and 200-nm diameters were determined by a high-sensitivity liquid in situ particle spectrometer. Selected colloid samples were further characterized by electron microscopy to evaluate colloid shapes, elemental compositions, and mineral phases. In the course of this work, we developed protocols for sample collection, storage, and filtering that minimized the effects of these factors on colloid analyses.

The instrument used to measure colloid concentrations and size distributions was a spectrometer manufactured by Particle Measuring System, Inc., Boulder, CO, Model HSLIS-S50. This instrument operates on the principle that light scattered by a liquid borne particle resident in a laser beam is directly proportional to the particle size. It uses laser illumination with a high-resolution optical system, collecting particle scattered light over the range of scattering angles $\pm 40^\circ$. Sizing is accomplished in situ with pulse-height analysis of the light scattering. The instrument has a 50- to 200-nm size range. The laser used is a 30 mW solid-state device (780-nm wavelength). The optical system includes cylindrical reflective condensing elements and spherical reflective collecting optics. The 0.85 numerical aperture of collection objective optics provides a large solid angle for collecting particle scattering light. All glass optical elements are antireflective coated. The laser beam is focused to the sample region using astigmatic condensing optics. Particles in the sample area reflect and obscure the laser light. Some sensors detect particles by reflected light, others by obstruction. The size of a particle is determined from the amount of reflected or obscured light. Optics focus light onto a photodiode detector that transmits electrical pulses to a pulse-height analyzer board for analysis. The board sizes the particles into 16 classes so that reported sizes are 50–60 nm, 60–70 nm, and so on, up to 190–200 nm and > 200 nm. Instrument measurements of colloid size and concentration were downloaded to a computer using the spreadsheet program Excel. Sample dilution and injection rates were incorporated in the calculations using the Excel files. Total colloid concentration between the 50- and 200-nm size range was determined by adding measurements from all 16 channels. The final results of colloid particle counts versus colloid particle sizes were computer plotted using the program Cricket Graph.

A scanning electron microscope (JEOL, 6300-FXV) with the PGT EDS system was used for imaging and elemental analysis. A Philips CM-30 Analytical Electron Microscope equipped for x-ray microanalysis, electron diffraction, and conventional imaging was used for imaging colloid sizes, shapes, and diffraction patterns and for

producing elemental composition analyses. A JEOL 3000 field emission, high-resolution transmission electron microscope was used to image colloid shapes and lattice fringes.

A. PRELIMINARY STUDIES

The first set of HRMP samples we received consisted of groundwater from the ER-20-5 #1 well that had been filtered through a series of Millipore filters (pore sizes 50 nm and 100,000 nominal molecular weight).²¹ Each sample had been prepared at full strength, at 1:10 dilution, and at 1:100 dilution. The spectrometer detected an abundance of particles in the 50- to 200-nm size range in all the samples. Furthermore, the diluted samples contained more colloids than the undiluted. This result led us to investigate the colloid content of the source of “nanopure” water used for the dilutions and, ultimately, to examine a number of other sources of such water in use at our laboratory. We learned that the output of nanopure water systems may vary considerably in colloid content, and if the water is then stored, the container may contribute markedly to the colloid population. The magnitude of this problem in our laboratory is illustrated in Figures 1 and 2, which compare water from our system (in TA-48-28-101), from the storage bottle there, and from the cleanest water available (TA-48-1-304). It was apparent that our source of dilution water for this suite of samples contaminated them with colloids. It also appeared that even the undiluted water samples contained much colloidal material that should have been removed in the filtration process according to the advertised pore sizes of the filters.

Figure 1. Colloid size distribution for nanopure water samples.

Figure 2. Total colloid concentrations for nanopure water samples.

Our experiences with the filtered samples from ER-20-5 led us to examine our procedures for collecting, storing, and filtering water samples to see if these operations introduced extraneous colloids. For these studies, we used water from the J-13 well at the NTS because it was readily available to us and because its relatively low colloidal content made it suitable for particle analysis without having to dilute the samples. The J-13 water is similar to other NTS groundwater in having low ionic strength, a property important in colloid stability. Since many of our studies of groundwater involve filtration, we surveyed a number of types of filters as potential generators of contaminating colloids. We evaluated filters made of cellulose acetate, nylon, cellulose nitrate, cellulose esters, polyethersulfone, and polycarbonate. We discovered that all these filters may generate colloids in filtrates unless they have been thoroughly cleaned before use. We found that nylon filters washed three times with low-colloid nanopure water produced the least number of contaminating colloids, so this type of filter was used in subsequent groundwater studies.

Many of our groundwater studies at the NTS require rather large sample volumes, so we routinely use 208-L polyethylene drums for sample collection. These drums are potential sources of contaminant colloids. We compared J-13 water collected in a polyethylene drum with water collected at the same time in a precleaned Teflon bottle. Data shown in Figure 3 indicate that the drum added a very significant number of colloids to the water, especially in sizes below about 110 nm.

Figure 3. Colloid size distribution for J-13 water stored in different type of containers.

We filtered some J-13 water through precleaned nylon filters of nominal pore sizes of 450 and 200 nm and compared the filtrates with the unfiltered water. The results, as shown in Figures 4 and 5, were that more than half of the colloids in the size range 50 to 200 nm were removed by the 450-nm filter and over 70% by the 200-nm filter. It is clear that the process of filtering radically depleted the colloid population, even though the colloid sizes were much smaller than the nominal pore size of the filters. The colloid removal was significant over the entire range of colloid sizes that we could measure.

Figure 4. Total colloid concentration in J-13 water samples with and without filtration through 450-nm or 200-nm nylon filters.

Figure 5. Colloid size distribution for J-13 water stored in a 200-liter plastic barrel, filtered through a 450-nm or a 200-nm nylon filter.

There is often a period of weeks or months between the time water is collected in a 208-L drum and the time it is analyzed. This time lag usually presents no problems for radionuclide analyses but may not be suitable if colloids are to be analyzed. We did several investigations of the effects of both short-term (up to nine days) and long-term storage (190 days) on colloids in J-13 water. We monitored the changes that occurred in the water chemistry during nine days after a groundwater sample collected at the NTS was placed in a Teflon bottle open to the atmosphere in our laboratory. For our sample, the Eh increased from 210 mv to 293 mv, the dissolved oxygen from 5.2 mg/L to 6.7 mg/L, and the pH from 7.2 to 8.3. The sample was absorbing oxygen and releasing CO₂. We observed a marked increase in colloid concentration over this time period, especially in the colloids sized 100 nm to 200 nm. The sample stored in a polyethylene drum for 190 days showed a three-fold increase in total colloid content; increases were observed over all the size ranges. Interestingly, a sample stored over the same period in a capped Teflon container had a decrease in total colloid concentration.

If our observations of colloid behavior in J-13 water are applicable to other NTS groundwater in which the chemistry may be somewhat different and the colloid content higher, we must be aware of a number of problems in sampling. Samples analyzed immediately in the field may not be affected by alterations associated with storage containers and by water chemistry changes. However, the sophisticated equipment and environmental controls needed for accurate analyses are not generally available in a field situation. Filtering, whether done in the laboratory or the field, may not effectively separate colloids according to size. We observe small colloids being removed by large pore filters, and colloids of many sizes in filtrates passed through filters with nominal pore sizes that should have stopped them. No doubt excessive loading of these filters may have affected their performance. This problem is a particularly difficult one for us since we are seeking to analyze materials in extremely low concentrations and must deal with large volumes of groundwater. The filters themselves must be rigorously cleaned before use, and so must the sample containers. Even so, storage of water samples may produce radical changes in the colloid sizes and population. As a result of our preliminary studies, we have developed protocols for field and laboratory handling of NTS water samples, but it is clear that careful interpretation of data is required to avoid misunderstandings originating in colloid behavior after the sample is removed from its natural environment.

B. ER-20-5 #1 SAMPLES

In April 1998, water samples were collected during the pumping of ER-20-5 #1. These samples consisted of unfiltered water and water filtered through a 450-nm nylon filter. The colloid size distribution and total colloid concentration were analyzed within 24 hours after collection. The filter acquired a yellowish color and was certainly excessively loaded. Figure 6 shows that the number of colloids in the size range >200 nm is less than 1% of the total and that the concentrations increase as colloid size decreases. The concentration of colloids (50 to 200 nm) in the filtrate was about two orders of magnitude lower than in the unfiltered water, so it is apparent that many colloids in this size range were collected on the filter.

Figure 6. Colloid size distribution in ER-20-5 #1 water.

We compared the colloid concentration and size distributions for unfiltered samples from ER-20-5 #1 and #3 wells. These are shown in Figures 7 and 8. The concentrations and size distributions were rather similar.

Figure 7. Colloid size distribution for ER-20-5 #1 and #3 waters.

Figure 8. Total colloid concentration for ER-20-5 wells #1 and #3.

Colloidal material collected on the 450-nm filter was examined using both the scanning and transmission electron microscopes described earlier in section III. The elemental analysis produced from the scanning electron microscope indicated that components normal for silicates were present (Si, Ca, K, Al). The transmission electron microscope clearly showed colloids with a variety of sizes and shapes as shown in Figure 9. In previous studies, we have attempted to characterize colloid sizes in groundwater from ER-20-5 using serial filtration.²¹ It is apparent that separation of colloids by this technique was probably incomplete, and our data should be interpreted with this understanding in mind.

C. CHESHIRE SAMPLES

Two water samples from the Cheshire site (U-20n ps1 ddh) were analyzed for their colloid concentrations and size distributions. The sample from the upper transmissive zone around 812 to 913 m slant depth was collected in July 1998; that from the lower zone at 1244 to 1253 m slant depth was collected in September 1998. Our data are shown in Figures 10 and 11. The colloid size distributions from the two zones appear to be rather similar, but the lower zone has about 20 times more colloids. We believe that both these zones have been pumped enough to remove any particulates associated with drilling, but we do not know if these concentrations would change with further pumping. We plan to explore the question of whether the higher concentrations of radionuclides in the lower zone are related to the higher colloid concentration there.

Figure 9. Transmission electron micrograph of colloids from ER-20-5 #1.

Figure 10. Colloid size distribution in groundwater samples from Cheshire.

Figure 11. Total colloid concentration in groundwater samples from Cheshire.

IV. PROGRAM SUPPORT ACTIVITIES

A. DOCUMENT REVIEW

One of the important functions of HRMP personnel is to serve as expert reviewers of documents prepared by DOE/NV contractors concerning NTS programs. During the past year, Los Alamos personnel critically reviewed the draft of "Value of Information Analysis for Corrective Action Unit Nos. 101 and 102: Central and Western Pahute Mesa, Nevada Test Site, Nevada," prepared by IT. This document described a method for evaluating the critical data required to model radionuclide transport in groundwater in one region of the NTS. We also reviewed a draft of the "Western Pahute Mesa-Oasis Valley Hydrogeologic Investigation Wells Drilling and Completion Criteria," which set forth the methodology for drilling and completing the wells to be drilled in Oasis Valley. LLNL produced a document titled "Evaluation of the Hydrologic Source Term from Underground Nuclear Tests in Frenchman Flat at the Nevada Test Site" that described an approach for modeling radionuclide movement in that part of the NTS. We reviewed this paper and suggested several modifications. Shortly afterward, we received a document from IT titled "Draft Groundwater Data Documentation Package for the Frenchman Flat Corrective Action Unit" that was concerned with some of the same issues, and we were able to respond with comments similar to those for the LLNL paper. Our basic concern is that the Cambrian event in Frenchman Flat may not be representative of other tests in that area and thus should not be the principal source for hydrologic source term data.

B. SHORT COURSE ON RADIONUCLIDE MIGRATION

In 1996, our HRMP manager, Doug Duncan, asked us to design and deliver a one-day course for DOE personnel acquainting them with groundwater transport of radioactive contaminants at the Nevada Test Site. This short course was first offered in 1997. We modified it somewhat for 1998 and videotaped the presentations as they were made. These videotapes have been duplicated and are available for use by HRMP or DOE presenters who need background information for technical talks or are engaged in public outreach meetings. The major topics covered are the nuclear test program; nuclear test phenomenology; source terms—radiologic and hydrologic; field experiments at the NTS; hydrogeologic controls; health effects and risk assessment; and corrective action and monitoring strategies for the NTS. We hope this course will prove to be a valuable resource for those needing to present information on NTS activities.

C. WORKSHOP/CONSULTATION

In July 1998, we participated in a two-day DOE/NV workshop sponsored by the Yucca Mountain Project and the Environmental Restoration program on the subjects "anthropogenic analogues" and "colloid facilitated transport." The objective of the workshop was to produce a summary of the information from the Hanford site, the Idaho National Engineering and Environmental Laboratory, and the NTS that is applicable to Yucca Mountain models of vadose zone flow and transport and will be used for planning purposes. We described our observations of plutonium migration at the Tybo-Benham site and discussed its relevance to the Yucca Mountain Project. In our

role as providers of technical expertise, we were able to assist the DOE/NV manager for Environmental Restoration in answering some queries from the National Academy of Sciences concerning radionuclide migration at the NTS. This group from the Academy was concerned with remediation of buried and tank wastes.

D. PUBLICATIONS

Publications prepared during the past year include our FY 1997 annual report²¹ on our HRMP activities, a comprehensive review article⁴⁵ on the Cheshire (U-20n) site and the studies conducted there since 1976, and a report³⁹ in an international journal on our observations of plutonium migration at the Tybo-Benham site.

ACKNOWLEDGMENTS

We appreciate the assistance of our colleagues at LLNL, DRI, USGS, IT, and BN in sample collection and the contributions of fellow workers at LANL in sample analyses. N. Goldman provided editorial assistance on this report. Our work was made possible by the financial support of two divisions at DOE/NV: Defense Programs and Environmental Restoration.

REFERENCES

1. D. C. Hoffman, "Summary of RNM Activities During FY-76 and FY-77 (July 1, 1976 to September 30, 1977)," Los Alamos Scientific Laboratory memorandum CNC-11 to R. W. Newman (November 10, 1977).
2. W. R. Daniels, "Summary of RNM Activities During FY-78 (October 1, 1977 to September 30, 1978)," Los Alamos Scientific Laboratory memorandum CNC-11 to R. W. Newman (November 16, 1978).
3. W. R. Daniels, "Summary of RNM Activities During FY-79 (October 1, 1978 to September 30, 1979)," Los Alamos Scientific Laboratory memorandum RNM-CNC-11/11/79-2 to R. W. Newman (November 9, 1979).
4. W. R. Daniels, Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1979 — September 30, 1980," Los Alamos National Laboratory report LA-8670-PR (February 1981).
5. W. R. Daniels, Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1980 — September 30, 1981," Los Alamos National Laboratory report LA-9192-PR (February 1982).
6. W. R. Daniels, Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1981 — September 30, 1982," Los Alamos National Laboratory report LA-9691-PR (May 1983).

7. W. R. Daniels and J. L. Thompson, Comps. and Eds., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1982 — September 30, 1983," Los Alamos National Laboratory report LA-10121-PR (April 1984).
8. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1983 — September 30, 1984," Los Alamos National Laboratory report LA-10372-PR (February 1985).
9. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1984 — September 30, 1985," Los Alamos National Laboratory report LA-10644-PR (January 1986).
10. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1985 — September 30, 1986," Los Alamos National Laboratory report LA-11081-PR (August 1987).
11. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1986 — September 30, 1987," Los Alamos National Laboratory report LA-11223-PR (February 1988).
12. J. L. Thompson, Comp. and Ed., "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).
13. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Hydrology/ Radionuclide Migration Project, October 1, 1988 — September 30, 1989," Los Alamos National Laboratory report LA-11947-PR (November 1990).
14. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Hydrology/ Radionuclide Migration Project, October 1, 1989 — September 30, 1990," Los Alamos National Laboratory report LA-12100-PR (May 1991).
15. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Hydrology/ Radionuclide Migration Project, October 1, 1990 — September 30, 1991," Los Alamos National Laboratory report LA-12338-PR (June 1992).
16. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Hydrology/ Radionuclide Migration Project, October 1, 1991 — September 30, 1992," Los Alamos National Laboratory report LA-12498-PR (January 1993).

17. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Hydrologic Resources Management Program, October 1, 1992 — September 30, 1993," Los Alamos National Laboratory report LA-12721-PR (January 1994).
18. J. L. Thompson, Ed., "Laboratory and Field Studies Related to the Hydrologic Resources Management Program, October 1, 1993 — September 30, 1994," Los Alamos National Laboratory report LA-12917-PR (March 1995).
19. J. L. Thompson, "Laboratory and Field Studies Related to the Hydrologic Resources Management Program, October 1, 1994 — September 30, 1995," Los Alamos National Laboratory report LA-13064-PR (November 1995).
20. J. L. Thompson, "Laboratory and Field Studies Related to the Hydrologic Resources Management Program, October 1, 1995 — September 30, 1996," Los Alamos National Laboratory report LA-13270-PR (March 1997).
21. J. L. Thompson, "Laboratory and Field Studies Related to Radionuclide Migration at the Nevada Test Site, October 1, 1996 — September 30, 1997," Los Alamos National Laboratory report LA-13419-PR (February 1998).
22. D. C. Hoffman, J. R. Stone, and W. W. Dudley, Jr., "Radioactivity in the Underground Environment of the Cambric Nuclear Explosion at the Nevada Test Site," Los Alamos Scientific Laboratory report LA-6877-MS (July 1977).
23. J. L. Thompson, Comp. and Ed., "Interim Report of the Drilling History and Analysis for Nevada Test Site Hole UE3e#4," Los Alamos National Laboratory report LA-12042-MS (December 1990).
24. E. A. Bryant, "Colloid Research at the Nevada Test Site," Los Alamos National Laboratory report LA-12301-MS (May 1992).
25. E. A. Bryant, "The Cambric Migration Experiment — A Summary Report," Los Alamos National Laboratory report LA-12335-MS (September 1992).
26. G. J. Nimz and J. L. Thompson, "Underground Radionuclide Migration at the Nevada Test Site," US Department of Energy/Nevada Field Office report DOE/NV-346 (November 1992).
27. D. K. Smith, B. K. Esser and J. L. Thompson, "Uncertainties Associated with the Definition of a Hydrologic Source Term for the Nevada Test Site," Lawrence Livermore National Laboratory report UCRL-ID-120322 (May 1995).

28. D. C. Hoffman, "A Field Study in Radionuclide Migration," in *Radioactive Waste in Geologic Storage*, S. Fried, Ed., ACS Symposium Series 100 (American Chemical Society, Washington, DC, 1979), p. 149.
29. D. C. Hoffman, W. R. Daniels, K. Wolfsberg, J. L. Thompson, R. S. Rundberg, S. L. Fraser, and K. S. Daniels, "A Review of a Field Study of Radionuclide Migration from an Underground Nuclear Explosion," International Atomic Energy Agency report IAEA-CN-43/469 (1983).
30. D. C. Hoffman and W. R. Daniels, "Assessment of the Potential for Radionuclide Migration from a Nuclear Explosion Cavity," in *Groundwater Contamination, Studies in Geochemistry* (National Academy Press, Washington, DC, 1984), p. 139.
31. J. L. Thompson and J. S. Gilmore, "Migration of Fission Products at the Nevada Test Site: Detection with an Isotopic Tracer," *Radiochim. Acta* 52/53 229 (1991).
32. J. L. Thompson, "Radionuclide Migration Studies at the Nevada Test Site," *Radiochim. Acta* 54, 149 (1991).
33. J. L. Thompson, "Studies of Fission Product Movement in Tuffaceous Media," *Radiochim. Acta* 58/59 273-276 (1992).
34. M. Mathews, K. Hahn, J. Thompson, L. Gadeken and W. Madigan, "Dalhart Post Shot Investigation," in *Proceedings of the Fourth International Symposium of the Minerals and Geotechnical Logging Society*, Toronto, Canada, August, 1991; published by the Society of Professional Well Log Analysts, 6001 Gulf Freeway, Houston, TX 77023 (1993).
35. W. L. Hawkins, D. A. Trudeau and S. L. Drellack, Jr., "Hydrogeologic Investigations at the Nevada Test Site," in *Proceedings of Industrial and Agricultural Impacts on the Hydrogeologic Environment*, Second USA/CIS Joint Conference on Environmental Hydrology and Hydrogeology: American Institute of Hydrology, May 17-20, 1993; Washington, DC., pp. 87-104.
36. J. L. Thompson, "Fission Product Distribution in Nuclear Test Cavities/Chimneys," in *Proceedings of the Fourth Annual International Conference on High Level Radioactive Waste Management*, Vol. 2, p. 1517, April 26-30, 1993, Las Vegas, NV (published by the American Nuclear Society and the American Society of Civil Engineers).
37. J. L. Thompson, "The Nevada Test Site: An Analog for a Nuclear Repository," in *Proceedings of the Fourth International Conference on the Chemistry and Migration Behavior of Actinides and Fission Products in the Geosphere*, p. 579, December 12-17, 1993, Charleston, SC (published by R. Oldenbourg Verlag, Munchen, 1994).

38. M. Mathews, K. Hahn, J. Thompson, L. Gadenken and W. Madigan, "Subsurface Radionuclide Investigation of a Nuclear Test," *J. Appl. Geophysics* **32** 279-291 (1994).
39. A. B. Kersting, et al., "Migration of Plutonium in Ground Water at the Nevada Test Site," *Nature* **397** 56-59 (1999).
40. Bechtel Nevada, "Hydrogeologic Characterization of U-3bh Collapse Zone" report prepared for US Department of Energy, Nevada Operations Office, August 1996 (DRAFT).
41. Bechtel Nevada, "Hydrogeologic Characterization of U-3at Collapse Zone: Data Report," report prepared for US Department of Energy, Nevada Operations Office, January 1997 (DRAFT).
42. Bechtel Nevada, "Hydrogeologic Characterization of U-3bl Collapse Zone" report prepared for US Department of Energy, Nevada Operations Office, November 1996 (DRAFT).
43. R. W. Buddemeier and D. Isherwood, "Radionuclide Migration Project 1984 Progress Report," Lawrence Livermore National Laboratory report UCRL-53628 (April 1985).
44. R. W. Buddemeier, "Hydrology and Radionuclide Migration Program 1985-1986 Progress Report," Lawrence Livermore National Laboratory report UCRL-53779 (September 1988).
45. D. A. Sawyer, J. L. Thompson, and D. K. Smith, "The Cheshire Migration Experiment," Los Alamos National Laboratory report LA-13555-MS (in review).