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Radionuclide Migration Project 1984 Progress Report

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April 1985

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Radionuclide Migration Project 1984 Progress Report

Abstract

This report presents previously unpublished results from the Lawrence Livermore National Laboratory's participation in the Radionuclide Migration Project at the Nevada Test Site (NTS). The report discusses the hydrogeologic settings and histories of studies associated with the Cheshire (U20n), Cambric (U5e), Nash (UE2ce), Bilby (U3cn), Bourbon (U7n), and Faultless (UC1) Events. Radionuclide and some chemical data are presented for water samples from cavity or chimney wells associated with the Cheshire, Cambric, and Bilby Events, and from satellite wells at the Cambric, Nash, Bilby, Bourbon, and Faultless Event sites.

The report also gives the results of studies of specific sampling or analytical methodologies. These studies demonstrated that the apparent migration of ¹⁵⁵Eu is an artifact of spectrometric misidentification of gamma- and x-ray peaks from other constituents. A potential problem with atmospheric contamination of samples collected with evacuated thief samples was also identified. Ultrafiltration techniques were applied to some of the Cheshire cavity samples collected, and preliminary results suggest that substantial amounts of activity may be associated with colloidal particles in the size range of 0.006 to 0.45 μ m.

A study has begun of the recharge of effluent water from RNM-2S (Cambric satellite well) into the desert floor as a result of nine years of continuous pumping. This report gives the initial results of unsaturated zone studies showing the propagation of moisture and tritium fronts through the shallow soil.

Geochemical modeling of the behavior of ruthenium and technetium was carried out, with particular emphasis on the identification of ionic species that would be potentially mobile under NTS ground-water conditions. The report compares the results with observations of ruthenium migration to the Cambric satellite well.

1. Introduction

This report presents the results of technical studies conducted by the Lawrence Livermore National Laboratory (LLNL) as part of the Radionuclide Migration (RNM) Project at the Nevada Test Site (NTS). The RNM Project is intended to assess the potential for radionuclide migration away from the underground nuclear test cavities at NTS, with particular emphasis on issues relating to ground-water contamination and transport. Figure 1-1 shows the locations of the sites and wells studied at NTS.

The project, which was initiated in 1974, continues as a multi-agency research project [LLNL, Los Alamos National Laboratory (LANL), the Desert Research Institute (DRI) of the University of Nevada, and the U.S. Geological Survey (USGS)] coordinated by the Nevada Operations Office of the U.S. Department of Energy (DOE-NVOO).

The agencies involved in the project have issued a variety of letter reports, technical reports, and scientific publications on aspects of RNM studies. LLNL has published a variety of topical reports,¹⁻¹⁶ but like other agencies involved has documented some of the results of ongoing studies in the form of letter reports and informal presentations.

The present report is intended to accomplish three objectives:

1. It is a comprehensive annual report of LLNL activities and results for the RNM project during FY84.

2. It summarizes the results of LLNL studies done in previous fiscal years that have not appeared in formal technical reports.

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3. It provides basic background information and a compilation of data reported by other agencies for ongoing studies for which no comprehensive or recent report exists.

The report is organized on a topical basis. Section 2 summarizes the results of field studies of NTS wells selected for study of possible radionuclide migration. Section 3 reports the results of field and laboratory experiments designed to evaluate or improve sampling and analytical techniques and to resolve specific questions of data interpretation. Section 4 describes a program initiated to use the ongoing Cambric pumping experiment to study recharge and tritium transport phenomena in an alluvial medium. Section 5 presents the results of geochemical modeling of ruthenium and technetium mobility in the context of the Cambric pumping experiment.

In addition to topical discussions and interpretations, the status of the various studies (completed or continuing) is indicated. Because this is an update report rather than a comprehensive review, the depth of treatment of the RNM well studies varies, depending on the availability of prior reports.

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Figure 1-1. Locations of experimental sites described in this report. Small numbers indicate NTS areas.

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2.1. General

2.1.1. Sampling and Blanks

(Contributors: J. Rego and R. W. Buddemeier)

This section describes the sample collection, processing, and analytical methods generally used for the RNM well samples. Unique or highly specific methods are described in the appropriate sections.

One of the mainstays of the LLNL RNM field program has been low-level gamma spectrometry of the salts from evaporated large-volume water samples. This approach provides high sensitivity for a variety of gamma-emitting nuclides of interest. Because it is more time-efficient than radiochemical separation, it permits us to screen substantial numbers of samples for evidence of radionuclide migration.

Two different approaches to sample evaporation are employed. One method consists of field collection of samples in one or more new plasticlined 55-gal drums. These drums are forwarded to LLNL, where the water is evaporated by addition of successive aliquots to stainless-steel drying pans in a specially designed oven located in a building remote from any locations where active handling of radioactive materials is conducted. The evaporated salts are composited and homogenized by hand, then packed into standard sample containers for gamma counting. These water samples are generally unfiltered, but have on occasion been filtered in the laboratory through 0.45- μ m Millepore or Nuclepore filters. In the data tables in this and the following sections, these samples are identified by the term "filter used." Those with no filter indicated or a filter size other than $1.0-\mu m$ are laboratory-evaporated samples, unless otherwise specified in the text. To minimize the possibilities of contamination, the drums are not reused, and the evaporation trays are thoroughly decontaminated between samples and are segregated according to whether they have been used for high- or low-activity samples.

The other evaporation system used is a field evaporator with a metered flow input through a $1.0-\mu$ m cartridge filter. The evaporator is a stainless-steel tank heated by gas-fired burners. The collected salt slurry is removed by hand and returned to LLNL in a stainless steel container for final oven drying, homogenization, packaging, and counting. The field sampler is flushed and decontaminated between samples and is used only for low-activity (satellite well) samples to minimize the possibility of cross-contamination. The tabulated data identified as results of a sample passed through a $1.0-\mu$ m filter are field evaporator samples, unless otherwise noted.

In determining the extremely low levels of activity present in some of the RNM satellite wells, the issue of an appropriate blank or background value becomes extremely important. The problems of establishing the filter blanks and of assigning observed activity to either dissolved or particulate fractions is discussed in some detail in Section 3.3.

Although scrupulous care is taken in correcting for detection system backgrounds and in processing the samples, the potential for sample contamination is always present, especially in view of the known surface contamination at NTS.

To obtain empirical estimates of a "field" blank, two large-volume water samples have been processed from water supply wells 5B and 5C, located in Frenchman Flat and completed to a depth of 230 m in the alluvial aquifer. The nearest detonation cavity is Cambric, which is 2.4 km upgradient. Because of the lack of significant transport observed in the Cambric pumping experiment, we have assumed that no subsurface contamination sources affect wells 5B and 5C.

The results of gamma analysis on the two evaporated large-volume water samples are presented in Table 2-1. The first, a laboratory-evaporated sample, showed no detectable activity; the second sample, collected using the field evaporator, indicated only a very low concentration of 137 Cs.

The level of ¹³⁷Cs seen is comparable to that observed in most of the satellite well samples, independent of the presence of any other nuclides. We believe that these measurements are real and that they represent unavoidable contamination from the ubiquitous surface ¹³⁷Cs at NTS. This is particularly true for field evaporator samples, which are somewhat more vulnerable to contamination by resuspended soil than are the drumcollected samples.

Any low-level counting evidence for radionuclide migration should be validated by repeated measurements, and ¹³⁷Cs in particular must be considered suspect at any concentration below several tenths of a dpm/L.

Sampling date	Solution vol (L) [salt wt (g)]	Filter				dpn	n/L (±	1 std dev	· %)				Ratio
		used	²² Na	⁴⁰ K	⁶⁰ Co	¹⁰⁶ Ru	¹²⁵ Sb	¹³⁷ Cs	¹⁵⁵ Eu	³ H	²³⁵ U	²³⁸ U	²³⁵ U/ ²³⁸ U
11/29/78	200.0 [158]	_	<0.002	19.25 (2.20%)	<0.002	<0.07	<0.02	<0.004	<0.004	NAª		4.33 (4.80%)	0.038
4/15/82	515.0 [182]	1.0 µm	< 0.002	17.60 (1.00%)	<0.002	<0.1	<0.01	0.04 (5.70%)	<0.004	NA	0.22 (1.50%)	2.88 (5.20%)	0.076

Table 2-1. Results of gamma analysis of well water at NTS wells 5B and 5C.

^aNA = Not analyzed.

2.1.2. Contract Analyses

(Contributors: R. Failor, J. Rego, and R. W. Buddemeier)

During 1983, aliquots of several archived salts from LLNL's large-volume RNM water samples were submitted to EAL Corporation (Richmond, Calif.) for low-level ⁹⁰Sr and plutonium analyses.

The archived salts were redried, mixed, and weighed; then weighed aliquots were taken for submission. One aliquot of Well 5B/5C salts was included as a blank; a mixture of reagent-grade salts approximating the composition of NTS ground water was prepared and used for substrates for submission of known activity blind standards. Measured volumes of ⁹⁰Sr and Pu standard solutions were distributed from a pipet over the surfaces of the salt samples, which were then dried and homogenized. EAL dissolved and analyzed the samples by standard techniques that have been shown to compare well with LLNL inhouse methods.¹ Because of the need to evaluate the sample data in the context of the blank and blind standard results, the data are presented here as a group in Table 2-2. Individually significant results are also included in relevant sections on specific wells.

Table 2-2 summarizes the results, correcting the decays to a reference time of July 1, 1983. Uncertainties are one standard deviation based on counting statistics only, and the weights are the oven-dried weights of the aliquot analyzed. The "equivalent volume" column gives the volume of the original water sample represented by the amount of salt analyzed, based on total volumes and salt weights for the original samples (see appropriate sections on individual wells). In assessing the results, it should be kept in mind that the original water samples were either not filtered or filtered only through a $1-\mu m$ cartridge, so that observed activities may be associated with suspended particles even though we treat them by convention as being dissolved.

On the basis of the $^{239/240}$ Pu data, we conclude that salt concentrations less than 10^{-3} pCi/g are indistinguishable from the blank, values in the $1-10 \times 10^{-3}$ range are indicative of the presence of Pu but quantitatively uncertain, and results in excess of 10^{-2} pCi/g are probably reasonably accurate. Based on this, none of the satellite wells shows any evidence of Pu, but positive results are definitely found for the Cambric cavity and probably for the Bilby chimney.

Evaluation of the ⁹⁰Sr data is somewhat more difficult. Most of the samples show apparent activity greater than that of the blank, but LANL has consistently reported 90 Sr \leq background in Cambric satellite samples, and no other radionuclides have been detected in the Bilby satellite (see Section 2.5), leading us to suspect that ⁹⁰Sr has not migrated to these wells. This finding, in conjunction with the relatively poor agreement for the spiked salt results and the Bilby satellite duplicates, indicates that about 10^{-1} pCi/g is probably the low limit for reliable results. This assumption leaves us with definite ⁹⁰Sr in the Cambric cavity (RNM-1) and the Nash satellite, and probably in the Bilby chimney as well. The other results must be considered inconclusive.

2.2. Cheshire (U20n)

(Contributors: R. W. Buddemeier, J. Rego, and J. Schweiger)

The Cheshire (U20n) Event was fired on February 14, 1976, with an announced yield in the 200- to 500-kt range. The site is on Pahute Mesa within the Silent Canyon Caldera formation. The device was detonated at a depth of 1167 m in a rhyolitic formation. The preshot water level was at a depth of 630 m.

The Cheshire site is of interest to the RNM program for three main reasons:

1. It is the only study site in tuff (as opposed to alluvium or carbonate aquifers).

					⁹⁰ Sr		²³⁹⁻²⁴⁰ Pu		
Sample No.	Source	Date	Salt wt (g)	Equiv vol (L)	$pCi/g (\pm 1 \text{ std dev \%})$	pCi/mL	pCi/g (± 1 std dev %)	pCi/mL	
1	Wells 5A/5B (blank)	4/15/82	132.5	375	$\frac{4.92 \times 10^{-3}}{(21.1\%)}$	1.74×10^{-6}	7.04×10^{-4} (20.9%)	2.49×10^{-9}	
2	Bilby chimney	10/21/81	45.7	134	1.66×10^{-1} (15.1%)	5.66 \times 10 ⁻⁵	6.90 × 10 ⁻³ (12.7%)	2.35×10^{-6}	
3	Bilby satellite	10/21/80	134.6	343	5.73 \times 10 ⁻³ (7.2%)	$2.25~\times~10^{-6}$	6.49×10^{-4} (25.2%)	2.55×10^{-7}	
9	Bilby satellite	10/21/80	138.6	353	1.63×10^{-2} (6.9%)	6.40×10^{-6}	5.43×10^{-4} (23.7%)	2.13×10^{-7}	
4	Bilby satellite	10/21/81	105.7	348	9.68 \times 10 ⁻³ (17.5%)	2.94×10^{-6}	6.96 × 10 ⁻⁴ (23.7%)	2.11×10^{-7}	
6	Nash satellite (UE2ce)	10/9/80	111.4	315	1.77 (1.5%)	6.26×10^{-4}	5.49 $ imes$ 10 ⁻⁴ (26.9%)	1.94×10^{-7}	
12	Nash satellite (UE2ce)	10/9/80	11 4.2	323	1.61 (1.9%)	$5.69~\times~10^{-4}$	3.14×10^{-4} (40.1%)	1.11×10^{-7}	
11	Nash satellite (UE2ce)	2/22/82	109.0	263	1.19 (2.1%)	4.93×10^{-4}	1.09×10^{-4} (75.1%)	4.52×10^{-8}	
7	Cambric cavity (RNM-1)	10/5/81	46.6	205	5.74×10^{1} (1.2%)	1.30×10^{-2}	1.34×10^{-2} (8.1%)	3.05×10^{-6}	
8	Cambric satellite (RNM-2S)	11/10/82	82.0	313	5.83×10^{-2} (15.1%)	1.53×10^{-5}	6.98 × 10 ⁻⁴ (31.4%)	1.83×10^{-7}	
					⁹⁰ Sr (pCi/	g)	²³⁹⁻²⁴⁰ Pu (pC	i/g)	
					Spiked	Reported	Spiked	Reported	
5	Spiked blank (blind standard)	_	99.1	_	1.93×10^{-2}	1.38×10^{-2} (10.2%)	1.82×10^{-3}	2.39×10^{-3} (13.6%)	
10	Spiked blank (blind standard)	_	49.3	_	3.87×10^{-1}	2.15×10^{-1} (2.9%).	3.65×10^{-2}	3.36×10^{-2} (4.8%)	

 Table 2-2.
 Results of sample analyses of 12 archived salts.

2. The detonation was more recent and larger than most of the other shots under investigation, thus providing a larger source term and a better opportunity to study the behavior of the short-lived radionuclides.

3. The site is only 8 km from the western boundary of NTS in a permeable formation with a surface water table gradient trending southwest, making it a likely candidate for relatively prompt off-site transport of radionuclides.

A re-entry hole (U20n PS 1DDH) was slant drilled during June and July 1976. The hole was sidewall cored and drilled to a total slant depth of 1378 m (vertical depth approximately 1287 m; estimated to be 60 m below cavity bottom). The casing was perforated from 1322- to 1321-m slant depth (approximate vertical interval: 1195-1220 m). A pump was installed at 1292-m slant depth (1206 m vertical), and a water sample was pumped. On pullout, the pump stuck at 610 m and could not be dislodged.² In March 1981, the pump was forced to the bottom of the hole and a bridge plug was set above it; the hole was then tested and found to be watertight. During July and August 1983 the hole was logged and scraped and the casing was perforated in the slant-depth interval between 1293 and 1306 m (vertical depth range 1206-1220 m). The well was surged, pressurized samples were taken by LANL, and a pump was installed. An initial attempt to pump water samples in August 1983 was aborted when the pump failed. After installation of a new pump at a slant depth of 922 m (vertical depth 861 m), the well was pumped between September 7 and 9, 1983. Teams from LANL, LLNL, USGS, and Reynolds Electrical and Engineering Co., Inc. (REECo) took samples and measurements during the pumping period. A total of 137,000 gal was pumped before the pump was shut down. Pumping and sampling were restarted on July 31, 1984; the results of this second test period will be reported later. REECo radionuclide analyses have been reported by letter.³ Table 2-3 summarizes the chronologic history of RNM studies, and Fig. 2-1 shows a vertical cross section of the experimental site.

2.2.1. Results

Although the initial sampling (1976) was intended to obtain water from below the lower boundary of the cavity in order to establish background activity levels, the samples contained ⁸⁹Sr, ⁹⁰Sr, ¹⁰³Ru, ¹⁰⁶Ru, ¹²⁴Sb, ¹²⁵Sb, ¹⁸¹W, ¹⁸⁸W, ⁸⁵Kr, ²³⁹Pu, and ³H (tritium levels reached 2000 nCi/mL), indicating that water had been drawn from or had already migrated below the cavity.^{4,5}

During the September 1983 sampling period, LLNL analyzed samples in the field for dissolved oxygen (DO), temperature, Eh, pH, and alkalinity by the methods described in Appendix A. Table 2-4 presents the results of these observations.

Date	Chronology
2/14/76	Detonation, hole U20n; yield range 200-500 kt. Working point 1167 m.
6/29/76	Hole U20n PS 1DDH drilled to 1287 m. Cased to 1284 m. Perforations 1239–1247; 1254–1259; 1267–1272.
9/13/76	Pump installed at 1227 m. Rate = 34 gal/min. Quit 45 min later.
9/15/76	Pump pulled.
9/24/76	Pump started down hole but stuck at 610 m (top).
9/29/76	Sandia camera employed downhole.
10/4/76	All attempts to recover stuck pump unsuccessful, abandoned until funding available.
3/20/81	Derelict pump pushed to the bottom of the hole; top of debris at 1229 m. Tubing filled with tagged water. Pressure tested 2-1/2 d.
5/20/81	Casing full of water; awaiting pump.
9/7/83	Perforations at 1207 and 1220 m; new pump installed at 861 m. Series of samples taken by LLNL and LANL for field and laboratory measurements. Shut down 9/9/83.
7/31/84	Pumping resumed.
8/1/84	LANL water collection. Rate 23 gal/min.
10/23/84	Large-volume samples taken by LLNL and LANL prior to shutdown.

Table 2-3. Cheshire (U20n) chronology.



Figure 2-1. Cheshire site cross section.

Time	Meter	DO	Temp	Eh (mV)		Alkalinity (p	opm CaCO ₃
(h)	(gal)	(ppm)	(°C)	(vs NHE) ^a	pН	NF ^b	F ^c
9/7/83							
1024	—	1.8	26.9	94.4	9.82		_
1050		0.3	32.5	- 53.7	9.91	—	_
1115	1898	3.5	34.2	238.5	8.29		<u> </u>
1146	3136	4.7	35.3	257.9	8.74	97	
1234	5372	4.4	36.7	204.4	8.51	—	—
1330	7982	4.1	—	—	8.43	—	_
1430	10739	4.9	_	271.1	8.46	—	
1530	13545	4.2	39.6	259.5	8.45	_	_
1630	16346	4.0	39.4	252.8	8.53	99	_
1730	19173	3.6	40.6	239.6	8.67	_	
2130	30288	2.8	41.2	235.0	8.0	_	_
2230	33100	3.3	_	_	8.13	_	_
2330	35917	3.5	40.3	237.0	8.05		—
9/8/83							
0030	38731	3.7	40.4	237.7	8.14		<u></u>
0130	41535	3.3	40.8	244.0	8.13	—	
0230	44351	3.2	—	—	8.52	—	
0330	47189	2.8	—	245.8	8.68	—	—
0430	50016	3.3	40.8	246.8	8.16	88	-
0530	52833	3.0	41.6	230.8	8.15	96	
0630	55631	3.0	40.2	231.3	8.05	—	_
0830	61187	3.0			8.57	—	—
0930	63802	3.3	40.2	231.3	8.15	88	_
1130	69466	2.8	40.8	234.5	8.65	—	_
1200	71064	2.7	42.8	263.0	8.65	96.5	94.5
1600	81372	2.8	44.0	254.3	8.68	118	98
2050	96660	3.1	44.0	302.7	8.69	110	103
9/9/83							
0025	107124	2.8	40.9	303.6	8.63	98	92
0500	120426	2.8	42.1	249.0	8.64	97.5	97
0833	130480	2.9	42.9	278.8	8.65	105	99

Table 2-4. Cheshire field measurements.

^a NHE = normal hydrogen electrode.

^b NF = not filtered.

^c $\mathbf{F} = \mathbf{filtered}$.

Samples were taken at intervals for laboratory chemical analysis. Samples for radionuclide analysis were taken beginning at corrected flowmeter readings (gal) of 56,590 (188-L sample); 71,603 (\sim 19-L sample); 81,370, 96,660, 107,125, 120,425, and 130,480 (all \sim 2-L samples); and 131,860 (200-L sample). All samples smelled of oil, and the final large-volume sample had a visible oil slick.

In the laboratory, the chemistry samples were $0.45-\mu$ m filtered and analyzed for cations by inductively coupled plasma spectrometry (ICP), conductivity, and titration alkalinity; for carbonate by autoanalyzer; and for other anions by ion chromatography. Table 2-5 presents the results of these analyses. Analyses were carried out in two sets. Samples labeled #1, #2, etc., were analyzed first and the results circulated to RNM participants in an informal letter report; after discussion of possible trends in the data, additional analyses were carried out to fill out the series. The analytical precision may be estimated from the replicate analyses of sample #6.

The small (2- and 19-L) samples were all field filtered through the same set of four 0.45- μ m Nuclepore filters. A 1-L aliquot of the 19-L sample was gamma counted in a Marinelli beaker, and all of the small samples were combined to yield a single 28-L composite sample. The 200-L sample collected at 131,860 gal was laboratory filtered $(0.45 \,\mu\text{m})$ and evaporated, and the salts were gamma counted. The 188-L sample taken at 56,590 gal and the 28-L composite sample were both ultrafiltered; procedures and results are discussed in detail in Section 3.3. Table 2-5 summarizes the results of filtrate analyses for both the "standard" (0.45- μ m filtered) and the ultrafiltered (~0.006- μ m) samples. Table 2-6 presents the total filterable activity $>0.45 \,\mu$ m (prefilter plus $0.45 \,\mu$ m filter) for each sample analyzed; ultrafiltration

					Ion cl	nromatograp	hy (IC), autoa	nalyzer (AA), and other	results			
		3,136 ^a		13,545		49,756		72,143		107,124		130,480	130,480
Analysis		#1	5860	#2	19,173	#3	56,590	#4	96,660	#5	120,376	#6	#6
Fluoride	(mg/L)	6.2	6.2	6.2	6.0	6.0	6.0	5.9	6.0	6.2	6.0	6.1	6.2
Chloride	(mg/L)	14.6	14.3	13.4	13.6	13.3	13.0	13.4	13.7	15. 2	13.3	13.4	14.4
Nitrate	(mg/L)	1.0	1.5	0.3	1.4	_	2.0	0.4	1.6	2.4	1.2	2.0	2.4
Sulfate	(mg/L)	34.35	33.9	33.5	32.8	32.8	33.4	32.8	33.8	35.2	33.7	34.4	35.8
Carbonate	(meq/L)	2.91	2.72	2.60	2.57	2.52	2.55	254	254	284	257	252	287
Conductivity	(mhos/cm)	403		395		380							
³ H	(µCi/mL)	0.406		0.402		0.400							
Alkalinity	(meq/L)	2.45		2.33		2.35							
						ICP a	analysis - filt	ered and aci	dified				
Element	Detection	3136		13,545		49,756		72,143		107,124		130,480	130,480
(mg/L)	limit	#1	5860	#2	19,173	#3	56,590	#4	96,660	#5	120,376	#6	#6
Aluminum	0.020	1.11	1.36	0.369	1.45	0.261	1.80	0.373	1.49	2.14	2.16	1.83	2.43
Boron	0.020	0.112	0.114	0.097	0.100	0.107	0.112	0.100	0.095	0.102	0.107	0.100	0.112
Cadmium	0.003	0.001	0.000	0	0	0.001	0.002	0.000	0.001	0	0.00	0.00	0.002
Cobalt	0.003	0.001	0	0	0	0	0	0	0	0	0	0.001	0
Copper	0.003	0	0	0	0	0.002	0	0.0	0	0	0		0
Iron	0.004	0.144	0.291	0.255	0.238	0.055	0.166	0.298	0.161	0.222	0.275	0.226	0.362
Lithium	0.001	0.255	0.189	0.174	0.177	0.175	0.162	0.161	0.154	0.169	0.169	0.163	0.159
Manganese	0.0005	0.011	0.022	0.036	0.087	0.073	0.096	0.041	0.107	0.056	0.126	0.105	0.110
Molybdenum	0.004	0.030	0.024	0.021	0.021	0.018	0.017	0.019	0.020	0.019	0.022	0.021	0.020
Nickel	0.008	0.022	0.022	0.044	0.006	0.001	0.004	0.006	0.004	0.0	0.003	0.001	0.006
Phosphorus	0.124	0.162	0.087	0.136	0.139	0.121	0.098	0.143	0.095	0.113	0.136	0.087	0.152
Lead	0.016	0.016	0.028	0.020	0.031	0.011	0.017	0.020	0.027	0.017	0.020	0.024	0.027
Selenium	0.100	0.047	0.065	0.061	0.042	0.057	0.055	0.063	0.042	0.048	0.055	0.054	0.057
Silicon	0.008	30.5	31.0	28.0	31.3	27.8	31.7	28.1	31.0	32.9	33.2	32.0	34.1
Strontium	0.012	0.070	0.031	0.014	0.017	0	0.006	0.005	0.007	0.0	0.0	0.003	0.006
Uranium	0.084												
Vanadium	0.003	0.003	0.003	0.001	0.001	0.000	0.001	0.002	0.002	0	0	0.003	0.002
Zinc	0.008	0.018	0.015	0.013	0.029	0.019	0.010	0.010	0.006	0.006	0.006	0.005	0.006
Calcium	0.020	8.64	4.63	3.17	3.17	2.09	2.03	1.71	1.64	1.77	1.73	1.59	1.57
Potassium	0.536	4.81	2.64	1.81	3.20	2.32	2.73	1.79	2.88	3.06	3.24	2.86	3.36
Magnesium	0.004	0.168	0.090	0.355	0.051	0.039	0.044	0.631	0.034	0.092	0.152	0.077	0.253
Sodium	0.024	85.1	74.8	72.5	80.9	80.5	72.2	71.8	69.7	81.3	81.9	73.9	72.2

 Table 2-5.
 Results of analyses of Cheshire samples.

^a Meter readings in gal (corrected); numbers denoted by # are sample numbers previously reported. Uncertainties: $CO_3^{=} = 3 \text{ meq/L}$; F, Cl, $NO_3 < 0.05 \text{ mg/L}$; SO₄⁼ = 0.05 mg/L. The last sample is a replicate of 130,480 gal.

	Field a	aliquot ^a	Barrel (131,860) ^b	Barrel (56,790) ^b
	0.45-μm	0.006-µm	0.45-μm	0.006-µm
Nuclide	28 L	28 L	200 L	188 L
²² Na	2.80	2.86	2.67	2.54
	(55%)	(26%)	(8.7%)	(20.8%)
⁴⁰ K	NL ^c	NL	8.45	3.87
			(6.8%)	(29.6%)
⁵⁴ Mn	NL	NL	0.38	<0.17
⁶⁰ Co	NL	NL	<0.1	
⁸⁵ Kr	35,100	NL	NL	NL
	(7%)			
¹⁰⁶ Ru	NL	NL	156.9	<7.7
(622 keV)			(1.7%)	
¹²⁵ Sb	1858	1709	2067	1703
	(4%)	(4%)	(0.6%)	(0.6%)
¹³⁴ Cs	4.26	2.44	3.83	2.77
	(2%)	(22%)	(5.2%)	(16%)
¹³⁷ Cs	4351	3596	5135	3238
	(2%)	(2.05%)	(0.7%)	(1.7%)
¹⁴⁴ Ce	NL	NL	25.82	<5.6
			(6.1%)	
¹⁵² Eu	NL	NL	14.47	<0.81
			(2.1%)	
¹⁵⁴ Eu	NL	NL	14.47	<0.85
			(2.1%)	
¹⁵⁵ Eu	NL	NL	18.91	<2.9
			(2.5%)	

Table 2-6. Cheshire sample radionuclide inventory: filtrate samples calculated to day 253.5 (1983). Units are dpm/L (\pm 1 std dev %).

^a Field aliquot = composite of samples; see text for volumes.

^b Corrected meter reading (gal) at time of sampling.

^c NL = not detected, no limit calculated.

retentate activities are presented and discussed in Section 3.3. Table 2-7 presents the total activity inventory for each sample; this is the sum of the activities of all filters, the ultrafilter retentate (if any), and the final filtrate. All activities are normalized to dpm/L. The uncertainties reported are based on counting statistics only and are unrealistically precise; as discussed in Appendix A, actual experimental uncertainties are probably in the range of \pm 20%.

2.2.2. Discussion

In both the early and the more recent sampling, there has been considerable concern about the degree to which the samples obtained actually represent formation water unaffected by well construction or conditioning. Chemical analyses were seen as one way to evaluate sample quality, especially because all drilling fluids had been tagged with lithium salts (\sim 20 ppm Li).

Figure 2-2 plots lithium concentrations as a function of volume pumped. There appears to be a calibration shift between the two sets of analyses, but both sets indicate an initial drop followed by stabilization after a total volume of

50,000–70,000 gal pumped. The pattern is seen even more clearly for calcium (possibly influenced by casing cement) in Fig. 2-3, which indicates stable concentrations after 70,000 gal. Table 2-3 shows that pH results show a similar pattern, stabilizing from 69,206 gal onward. The Cl⁻ and CO_3^- values in Table 2-4 show an initial decrease in the first two samples. Tritium values were stable throughout, as were most other chemical constituents.

Table 2-7 shows that the total activity (filters plus filtrates) inventories of the two large-volume samples are in reasonable agreement, indicating that there was no major shift in radionuclide content between the two sampling times. Values for the 28-L composite sample are similar but somewhat higher; Tables 2-6 and 2-7 show that for most of the nuclides the additional activity is apparently associated with the particulate phase (see the discussion in Section 3.3.).

Solution activity is dominated by 137 Cs and 125 Sb; nearly all of the 125 Sb and more than half of the 137 Cs (and 134 Cs) is not filterable even by ultra-filtration. There is good agreement between the two 0.45- μ m filtrates, and also between the two

	Field ali	quot ^a	Barrel (131,	,860 gal)	Barrel (56,330 gal)		
	Composite filter	Total filter +	Composite filter	Total filter +	Composite filter	Total filter +	
Nuclide	28 L	filtrate	200 L	filtrate	188 L	filtrate	
²² Na	1.12	3.9	0.32	2.99	<0.10	2.54	
	(30.6%)		(38%)				
⁴⁰ K	0.8	0.8	3.14	11.59	3.71	7.58	
	(32.6%)		(11.6%)		(19.4%)		
⁵⁴ Mn	0.87	0.87	0.25	0.63	0.56	0.56	
	(16%)		(31.4%)		(57%)		
⁶⁰ Co	NL ^b	7.32	2.06	5.38	4.57	4.57	
			(4.3%)		(3.0%)		
¹⁰⁶ Ru	222	222	40.8	197.7	150.6	150.6	
(622 keV)	(1.0%)		(4.9%)		(19.0%)		
¹²⁵ Sb	79.1	1937	17.1	2084	91.2	1794	
	(1.3%)		(3.4%)		(2.6%)		
¹³⁴ Cs	1.94	6.2	0.89	4.7	1.57	4.34	
	(8.0%)		(3.9%)		(20.5%)		
¹³⁷ Cs	3651	8002	1422.6	6557	2699	5937	
	(1.0%)		(0.9%)		(1.1%)		
¹⁴⁴ Ce	48.1	48.1	7.82	33.6	26.51	26.51	
	(4.7%)		(10.6%)		(20%)		
¹⁵² Eu	23.8	23.8	4.01	18.0	14.41	14.41	
	(1.9%)		(2.5%)		(19%)		
¹⁵⁴ Eu	30.8	30.8	5.4	19.87	19.81	19.81	
	(1.7%)		(2.2%)		(10.8%)		
¹⁵⁵ Eu	56.0	56.0	9.8	28.7	30.37	30.37	
	(5.2%)		(2.9%)		(5.4%)		
²⁴¹ Am	5.48	5.48		0.11	2.76	2.76	
	(55.1%)		NL		(50.8%)		

Table 2-7. Cheshire sample radionuclide inventory: total filter + filtrate inventory calculated to day 253.5 (1983). Units are dpm/L (\pm 1 std dev %).

^a Field aliquot = composite of samples, see text for volumes. For the field aliquot and the 56,330-gal barrel sample, the composite filter value includes both standard filters and the ultrafilter (0.006- μ m) retentate.

 $b \hat{N}L = not$ detected, no limit calculated.



Figure 2-2. U20n (Cheshire) lithium vs volume pumped.



Figure 2-3. U20n (Cheshire) calcium vs volume pumped.

0.006- μ m ultrafiltrates. This agreement, together with the similarities in large-volume total activity inventories discussed above, suggests that there is no evidence for any consistent relationship between sample activity and volume pumped. It should also be noted that there is good agreement on the salt contents in the <0.45- μ m size range for the two large-volume samples. The 200-L filtrate yielded 63.84 g when dried, or 0.32 g/L; the 188-L ultrafiltrate yielded 48.09 g and the 3-L retentate produced 11.18 g, for a combined total of 0.31 g/L. The retentate/ultrafiltrate salt concentration ratio is about 15, which is similar to but slightly higher than the chemical concentration ratios reported in Section 3.3.

The observations concerning the smaller but readily measurable concentrations of ⁵⁴Mn, ⁶⁰Co, ¹⁰⁶Ru, ¹⁴⁴Ce, and the europium isotopes are of particular interest. In the conventionally treated sample (0.45- μ m filter), significant concentrations are seen in the filtrate. The data of Table 2-6 show, however, that these nuclides are quantitatively removed by ultrafiltration in both of the other samples analyzed (although quantitative upper limits are not reported for the composite sample ultrafiltrate, they are well below the concentration levels reported for the 0.45- μ m filtrate). Because of the good agreement between the total activity inventories of the two large-volume samples, it seems safe to treat these samples as replicates, and the results therefore clearly imply that most of the transition and lanthanide element radionuclides are associated with particles in the 0.006- to $0.45 - \mu m$ size range.

These results are of considerable importance to calculations of distribution and retardation coefficients based on field data, and they further emphasize the need for study of the role of submicron particles in radionuclide transport processes.

Further analyses and filtration studies are planned for the U20n samples collected during 1984.

2.3. Cambric (U5e)

(Contributors: J. Rego and R. W. Buddemeier)

The RNM experiments associated with the Cambric site have been extensively documented both in technical reports⁶⁻¹⁴ and in publications in the scientific literature.¹⁵⁻²⁰ This report will be limited to the presentation of basic background material and previously unpublished LLNL data. Section 4 of this report describes a new experimental initiative based on the Cambric pumping experiment.

The Cambric (U5e) device was detonated on May 14, 1965, with a yield of 0.75 kt. The working point was at a depth of 294 m in alluvium; the alluvium-tuff contact was at a depth of 690 m and the preshot water table was 73 m above the working point. For a variety of reasons (yield, location, and aquifer type), the Cambric site was judged to be ideal for an active RNM experiment, and in 1974 two experimental holes were drilled.

A reentry hole, RNM-1, was slant drilled through the cavity on June 20, 1974, with core, water, and gas samples taken at selected intervals.10,18,19 After completion of the initial sampling, the packers between the two uppermost zones were drilled out and a pump was installed just above the cavity to permit periodic resampling of the source. Satellite well RNM-2S was drilled in April 1974. It was located 91 m from the Cambric cavity, was drilled to a total depth of 340 m, and was perforated at 340 and 316 m. A pump was installed at 300 m. Figure 2-4 shows the Cambric site vertical cross section. In an effort to induce radionuclide migration by imposing an artificial hydraulic gradient on the system, continuous pumping of RNM-2S at a rate of approximately 1 m³/min was begun in October 1975. In October 1977, a larger pump was installed and pumping was resumed at a rate of 2.3 m³/min. Tritium breakthrough was observed in RNM-2S in December 1977. The tritium concentration in the RNM-2S effluent reached a maximum in July 1980 and has been slowly decreasing since then.

Numerous samples have been taken by LANL (see cited references) and LLNL over the course of the pumping experiment. Tables 2-8 and 2-9 give the histories of the RNM-1 and RNM-2S holes. Table 2-10 presents the results of LLNL analyses of large-volume water samples from RNM-1; RNM-2S results are given in Table 2-11.

The Cambric source term and the significance of changes in RNM-1 activities have been discussed in detail in various LANL publications; these observations will not be repeated here. In addition to tritium, LANL has measured ⁸⁵Kr (which apparently moves conservatively with the tritium), ³⁶Cl, and ¹²⁹I in RNM-2S samples. The only other nuclide detected in RNM-2S samples has been ¹⁰⁶Ru¹⁵; the low levels of ¹³⁷Cs reported in Table 2-11 are of the magnitude normally associated with ubiquitous surface contamination and are not considered significant (see Section 2.1.2). Although the actual ¹⁰⁶Ru activities observed are quite low because of the short half-life, the data suggest that the ¹⁰⁶Ru activity correlates with the tritium activity, indicating conservative behavior.



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Figure 2-4. Cambric site cross section.

Date	Event							
5/14/65	Detonation; yield 75 kt, working point 294.3 m.							
6/20/74	RNM-1 (cavity well) drilled; total slant depth 397 m, total depth 370 m, cored							
6/74-8/75	Five hydrologic zones in the well isolated and sampled.							
10/4/77	Pump installed at 270.7 m MSD. (Ref. Hoffman, Memo 10/11/77, LANL.) First reentry, pumped 56.8 m ³ from zones IV and V.							
11/30/77	Second reentry, LLNL and LANL samples taken.							
9/4/79	LLNL sampling.							
10/5/81	LLNL sampling.							
4/1/82	LLNL sampling.							

Table 2-8. Cambric (RNM-1) chronology, and dates of LLNL sampling.

D-series samples (Zone III) 1 to 17 ranged from 7.6 to 8.5 \times 10 6 dpm/L.

E-series samples (Zone IV) 1.8 \times 10⁵ dpm/L; ¹³⁷Cs 735 dpm/L.

F-series samples (Zone V) 6.2 \times 10⁴ dpm/L; ¹³⁷Cs 780 to 66 dpm/L.

Table 2-9. Cambric satellite (RNM-2S) chronology, and dates of LLNL sampling.

Date	Event
4/74	RNM-2S drilled, 91 m from cavity, total depth 340 m.
10/75	Pump installed at 300 m. Pumping commenced at a rate of 0.95–1.14 m ³ /min. Pumped continuously except for brief periods for repair and maintenance.
10/77	Pump replaced; pumping resumed at a rate of 2.27 m ³ /min. Sampling continued.
12/27/77	3 H breakthrough at total volume 1.44 $ imes$ 10 ⁶ m ³ pumped.
11/29/78	LLNL large-volume, low-level sampling.
3/14/79	LLNL large-volume, low-level sampling.
8/29/79	LLNL large-volume, low-level sampling.
11/10/82	LLNL large-volume, low-level sampling.
4/19/84	Field measurements and LLNL large-volume low-level sampling.

This result appears to be at odds with observation of the Bilby chimney showing that ¹⁰⁶Ru decreased much more rapidly than tritium activity between 1977 and 1981 (see Table 2-21, Section 2.5). This apparent contradiction may be the result of a redox equilibrium that permits ruthenium to exist either as the mobile anionic form RuO_4^- or as the more readily sorbed cationic species, depending on the Eh/pH conditions in the ground-water system. The chemical and geochemical characteristics of ruthenium are discussed in more detail in Section 5.

One unresolved issue relating to the Cambric pumping experiment is the question of whether the pumped effluent, which is discharged into an unlined ditch near the wellhead, may have recharged the water table over the course of the experiment. If this has occurred, the possibility that RNM-2S pumping may be sampling some fraction of recycled water will have to be considered; this could complicate quantitative modeling of the transport of radionuclides from RNM-1 to RNM-2S. This possibility, which was one of the motivations for initiating the Cambric plume study, is discussed in Section 4.

2.4. Nash (UE2ce)

(Contributors: R. W. Buddemeier and J. Rego)

The Nash Event was detonated on January 19, 1967 with an announced yield in the 20- to 200-kt range. The working point was at a depth of 367.5 m. Chimney collapse occurred shortly after detonation.

Satellite well UE2ce was drilled on January 25, 1977; the target location was 183 m due south of U2ce. Pawloski²¹ reports the location as 55.6 m south of U2ce, but this is almost certainly

Sampling date	Solution vol (L) [salt wt (g)]	Filter					dpm/L (\pm 1 std d	ev %)				Ratio
		used	²² Na	⁴⁰ K	⁶⁰ Co	¹⁰⁶ Ru	¹²⁵ Sb	¹³⁷ Cs	¹⁵⁵ Eu	³ H	²³⁵ U	²³⁸ U	²³⁵ U/ ²³⁸ U
9/4/79	390.0 [207.3] ^a		<0.004	15.22 (1.60%)	<0.002	<0.27	0.26 (19.20%)	84.46 (0.90%)	<0.004	$3.20 \times 10^{6^{b}}$ (<1.0%)	0.32 (5.90%)	4.63	0.069
10/5/81	404.0 [92]	-	<0.004	11.30 (1.10%)	<0.002	<0.28	0.20 (14.00%)	37.40 (1.90%)	<0.004	1.37×10^5 (3.30%)	0.15 (15.0%)	2.79 (5.10%)	0.052
4/1/82	181.0 [42]		<0.004	13.26 (1.50%)	0.04 (16.10%)	<0.63	<0.07	37.10 (1.70%)	<0.004	$7.20 imes 10^{4^{C}}$ (<1.0%)	0.18 (10.40%)	3.82 (8.40%)	0.048

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Table 2-10. Analysis of Cambric cavity well RNM-1 calculated to day 135 (1979).

^a Not dry weight. ^b LLNL analysis.

^c LANL analysis.

Table 2-11. Analysis of Cambric satellite well (RNM-2S) calculated to day 135 (1979).

Sampling date, m ³	Solution vol (L) [salt	Filter					dpm/L (⊣	1 std dev	<i>7</i> %)				Ratio
pumped	[5unt wt (g)]	used	²² Na	⁴⁰ K	⁶⁰ Co	¹⁰⁶ Ru	¹²⁵ Sb	¹³⁷ Cs	¹⁵⁵ Eu	³ H	²³⁵ U	²³⁸ U	²³⁵ U/ ²³⁸ U
$\frac{11/29/78}{2.65 \times 10^6}$	200.0 [158]		<0.004	22.36 (1.80%)	<0.002	0.16 (45.00%)	<0.02	0.004 (45.00%)	<0.004	1.80 × 10 ^{6^a}	0.20 (5.9%)	5.55 (5.00%)	0.036
2/7/79 2.88×10^{6}	209.0 [121]	-	<0.004	16.13 (2.60%)	<0.002	0.16 (40.00%)	<0.02	<0.006	<0.004	$2.40~\times~10^{6^{a}}$	0.12 (6.90%)	2.49 (47.00%)	0.048
3/14/79 2.98 × 10 ⁶	200.0 [113]	—	<0.004	17.27 (2.20%)	<0.002	0.15 (31.00%)	<0.02	<0.006	<0.004	2.63 × 10 ^{6^a}	0.17 (4.90%)	3.55 (26.00%)	0.048
8/29/79 $3.49 imes 10^{6}$	714.0 [278]	1.0 µm	<0.004	22.92 (1.30%)	<0.002	0.31 (9.00%)	0.01 (36.00%)	0.01 (3.00%)	< 0.004	4.23 × 10 ^{6^a}	0.09 (4.00%)	1.90 (6.00%)	0.047
4/17/80 4.20×10^{6}	740.0 [339]	1.0 µm	<0.004	28.61 (1.00%)	<0.002	0.18 (21.00%)	<0.01	0.02 (27.00%)	<0.004	6.10 × 10 ^{6^a}	0.30 (3.20%)	7.37 (5.50%)	0.042
7/23/80 4.51×10^{6}	863.0 [243]	1.0 µm	<0.004	18.92 (1.20%)	<0.002	0.22 (21.00%)	<0.01	0.01 (12.50%)	<0.004	$6.24 imes 10^{6^{a}}$ (<1.0%)	0.19 (2.40%)	4.73 (3.80%)	0.041
7/22/81 5.65 $ imes$ 10 ⁶	420.0 [122]	1.0 µm	<0.004	15.63 (1.60%)	<0.002	<0.02	<0.01	0.03 (11.00%)	<0.004	$6.02 imes 10^{6}{}^{b}$ (<1.0%)	0.13 (3.30%)	2.81 (4.80%)	0.046
$\begin{array}{r} 11/10/82 \\ 5.99 \ \times \ 10^6 \end{array}$	450.0 [118]	1.0 µm	<0.004	17.98 (1.00%)	<0.002	<0.01	<0.01	0.02 (12.40%)	<0.004	$6.24 \times 10^{6^{b}}$ (1.73%)	0.15 (3.00%)	2.50 (4.60%)	0.058
4/19/84 8.94 × 10 ⁶	315.7 [94.6]	1.0 µm	<0.004	18.24 (1.10%)	<0.002	<0.02	<0.03	0.003	<0.004	$4.73 \times 10^{6^{b}}$ (1.50%)	0.18 (2.50%)	2.44 (7.60%)	0.075

^a Tritium values selected from Ref. 6.

^b LLNL analyzed.

in error; we have paced off the distance from UE2ce to the Nash crater rim as 100 yards, and we believe that the hole is indeed located approximately 600 ft south of U2ce ground zero.²² The well was drilled to a total depth of 503 m, cased to 495 m, cemented at 422 m, and perforated in the 422- to 495-m interval.²¹

Based on cores and geophysical logs, the site stratigraphy is alluvium from the surface to 126 m, tuff from 126 to 340 m, and Paleozoic carbonates below 340 m. Measurements of depth to water in UE2ce in 1977 yielded values ranging from 426 to 434 m,²¹ suggesting that the equilibrium water table is probably below the cavity. The most probable direction of natural ground-water flow at the Nash site is southeastward, but no rigorous local determinations of gradient have been made and differing interpretations of the regional data yield flow directions ranging from due east to due south.²³

Area 2 in the general vicinity of UE2ce is a heavily used section of NTS; several shots have been fired within a 1-km radius of the Nash Event, and a substantial number within a slightly larger radius. This suggests the possibility that the hydraulic effects of other detonations, including water table variations and temporary changes in the rate and direction of local ground-water flow, may need to be considered in assessing the hydrology of the Nash site.

Based on the foregoing, we can see that the detonation occurred in the upper Paleozoic, that the bottom of the cavity probably does not extend into the normally saturated zone, and that the satellite well (assuming integrity of casing and seal) produces water exclusively from the carbonate aquifer. Figure 2-5 shows the Nash site vertical cross section.

2.4.1. Sampling and Analysis

Pumping and sampling have been conducted intermittently since 1977; Table 2-12 is a chronologic summary of activities. Figure 2-6 summarizes tritium activities from all sources (LANL, REECo, and LLNL) as a function of total volume pumped, with the dates and major events noted. Large-volume samples were collected by LLNL on March 15, 1978, October 9, 1980, February 22, 1982, August 9, 1982, August 10, 1983 and April 18-19, 1984. All samples were evaporated and the salts counted by low-level gamma spectroscopy. The 1984 sample was also filtered and the filter analyzed. Table 2-13 presents the results of these determinations, with tritium results added for completeness. The apparent europium results from the GAMANAL spectral analysis are almost

Date	Chronology					
1/19/67	Detonation; yield range 20-200 kt; working point 367.6 m.					
1/25/77	Satellite well, UE2ce, drilled and cored; perforated at 422–495 m. Water level 434 m					
4/12/77	Well released for RNM studies.					
5/18/77	Pump installed at 486.5 m. Water level 426.1 m.					
5/19-20/77	Pump tests and sampling at intervals. Water level 432.2 m.					
5/21/77-8/10/77	Pumping intervals as directed.					
11/15/77	Start of continuous pumping.					
12/21/77	Pump stopped. Excessive drawdown.					
2/27/78	Start of continous pumping.					
3/15/78	LLNL large-volume sampling.					
3/24/78	Pump shut down after excessive drawdown coinciding with NTS event.					
8/28/80	Pump replaced. Rate of 46 L/min; pump intake at 475.4 m. Water level 434 m.					
2/24/82	LLNL collection; throttled back to 23 L/min. Start of continuous pumping.					
7/9/82	LLNL large-volume collection.					
8/10/83	USGS collection for LLNL.					
4/18/84	Field measurements and large-volume collection.					

Table 2-12. Nash satellite well (UE2ce) chronology.



Figure 2-5. Nash site cross section.

certainly spurious, probably caused by thorium interferences (see Section 3.1).

LANL has performed analyses for 85 Kr, 137 Cs, 3 H, and 90 Sr. The 85 Kr results from pressurized water samples taken in 1977 and 1978 clustered in the range of 2–3 dpm/L at T_0 ; 137 Cs was not detected. The results of 90 Sr determinations

by LANL and EAL/LLNL are summarized in Table 2-14. When probable chemical yields for the EAL determinations are considered, these results are in agreement with LANL's 1978 results.

Chemical analyses of UE2ce water have been performed by the USGS and by LLNL. The results are summarized in Table 2-15.

Sample date, m ³	Net vol (L) [salt	Filter					dpm/L	(± 1 std d	ev %)				Ratio
total	wt (g)]	used	²² Na	⁴⁰ K	⁶⁰ Co	¹⁰⁶ Ru	¹²⁵ Sb	¹³⁷ Cs	¹⁵⁵ Eu	³ H ^a	²³⁵ U	²³⁸ U	²³⁵ U/ ²³⁸ U
3/15/78 9,250	200.0 [121.8]	1.0 µm	0.68 (4.40%)	41.90 (2.10%)	<0.002	<0.2	<0.03	<0.01	0.12 (27.00%)	2.80×10^7	0.22 (10.20%)	4.17 (14.50%)	0.052
10/9/80 13,969	1775.0 [628]	1.0 µm	0.39 (12.76%)	23.50 (1.40%)	<0.002	<0.5	<0.02	0.02 (17.00%)	0.07 (21.70%)	NA ^b	0.15 (6.70%)	3.96 (2.60%)	0.038
2/22/82 18,717	643.0 [267]	1.0 µm	0.14 (26.00%)	32.35 (1.20%)	<0.002	<0.2	<0.02	0.04 (19.00%)	0.10 (25.70%)	4.80 × 10 ⁶ (1.50%)	0.15 (6.60%)	3.12 (15.90%)	0.049
7/9/82 22,681	734.0 [393]	1.0 µm	0.98 (5.30%)	40.90 (3.90%)	<0.002	<1.3	<0.02	0.03 (14.50%)	0.04 (45.00%)	4.40×10^{7} (<1%)	0.28 (16.30%)	2.13 (7.50%)	0.130
8/10/83 40,285	175.0 [77.36]	0.45 µm	1.11 (11.20%)	24.87 (2.60%)	0.31 (26.00%)	<0.06	<0.12	0.14 (16.40%)	0.36 (19.40%)	$3.92~\times~10^7$	0.11 (13.10%)	2.28 (7.60%)	0.050
4/18/84 40,542	314.0 [177.9]	1.0 µm	1.97 (2.08%)	40.81 (1.10%)	<0.009	<0.06	<0.09	0.02 (27.30%)	0.38 (13.7%)	7.28×10^{7} (<1.0%)	0.19 (8.20%)	4.15 (2.80%)	0.045

Table 2-13. Analysis of Nash satellite well (UE2ce) samples calculated to day 74 (1978).

^a Tritium values from various sources (LANL, REECo, LLNL). ^b NA = not available.

Table 2-14. ⁹⁰Sr in UE2ce water, calculated to 1/19/67.

Sample date	Volume	Activity (dpm/mL)	Laboratory (reference)
08/03/77	200 mL	≤Background	LANL (24)
08/04/77	200 mL	≤0.005	LANL (24)
09/29/77	200 mL	0.016	LANL (24)
11/28/77	200 mL	\leq Background	LANL (24)
11/29/77	200 mL	≤0.03	LANL (24)
12/09/77	200 mL	\leq Background	LANL (24)
03/12/78	500 mL	≤Background	LANL (25)
03/13/78	500 mL	≤Background	LANL (25)
03/23/78	500 mL	≤Background	LANL (25)
03/24/78	500 mL	≤Background	LANL (25)
08/10/78	500 mL	0.0089	LANL (26)
08/14/78	500 mL	0.012	LANL (26)
08/28/78	500 mL	0.015	LANL (26)
09/01/78	500 mL	0.014	LANL (26)
10/09/80 ^a	319 L	0.00090	EAL/LLNL ^c
02/22/82	263 L	0.00074	EAL/LLNL ^c
05/(2 + 11)/82 ^b		≤Background	LANL (27)
05/(18 + 26)/82 ^b		≤Background	LANL (27)
FY83		≤Background	LANL (28)

^a Values are averaged for two replicates. ^b Composite samples. ^c See Section 2.3. Results are not corrected for chemical yield.

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Figure 2-6. Nash (UE2ce) tritium history. Tritium values are calculated to March 15, 1978.

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	1977	1978	1983	1984
	USGS	USGS	LLNL	LLNL
Chemical	18 samples	10 samples	2 samples	3 samples
K +	23 (1%)	34 (13%)	22.1 (0.3%)	22.3 (2%)
Ca ^{+ 2}	88 (8%)	97 (13%)	70.9 (0.4%)	94.7 (0.3%)
Mg + 2	37 (4%)	44 (7%)	31.4 (0.0%)	39.9 (0.1%)
Mg ⁺² Sr ⁺²	—	—	—	0.27
Na ⁺	41 (2%)	51 (4%)	45.8 (0.2%)	62.7 (0.5%)
Li ⁺	—	—		0.17
CI-	38 (17%)	103 (26%)	49.2 (<0.05%)	108.8 (<0.5%)
F -	0.7 (0.1%)	0.7 (0.1%)	1.05 (<0.05%)	0.9 (<0.5%)
SO_{4}^{-2}	25 (3%)	32 (3%)	56.8 (0.05%)	32.6 (<0.5%)
HCO ₃	499 (33%)	444 (53%)	620 (3%)	619.0 (3%)
SiO2	81 (3%)	71 (8%)	67.7	69.1
pH	7.1 (0.2%)	6.9 (0.3%)	7.5 (0.1%)	6.6 (0,5%)
Temperature	34.1 (0.8%)	32.5 (4.1%)	NA	33.2 (1.0%)

Table 2-15.	Chemical	analysis	of ground-water samples from UE2ce.
Mean values	s are given	in units	of mg/L (\pm 1 std dev %).

2.4.2. Discussion

The results of the chemical and radiochemical analyses of UE2ce water are qualitatively consistent but quantitatively rather variable. ³H, ⁸⁵Kr, and ²²Na are definitely present, and there is an indication of very low levels of ⁹⁰Sr. ³H and ⁸⁵Kr are volatile products that may have been injected some distance into the formation beyond the cavity boundary, and ²²Na is probably the result of fast neutron (n,2n) reactions with natural ²³Na in the minerals surrounding the detonation. All three nuclides are relatively mobile, and their production and transport modes are such that their presence in the ground water does not necessarily indicate direct interaction of ground water with the cavity. ⁹⁰Sr, if attributable to the Nash detonation, might be more indicative of an aqueous transport pathway from the cavity to the satellite well; however, the observed levels are so low and variable that it is difficult to be certain that they are not caused by sample contamination, and ⁹⁰Sr can be produced by decay of volatile 90Br and 90Sr precursors.

Four main factors may contribute to the variations in sample chemistry and activity: (1) The 8/10/83 sample, which shows the highest and most varied inventory of radionuclides, was the only one filtered through Nuclepore filters. Because LANL has shown that these filters contribute significantly to the blank levels of solutions passed through them (see Section 3.3.), we discount the significance of those nuclide results that are substantially different from the preceding and following samples. (2) The pumping at UE2ce has been intermittent and the rates variable. (3) The Paleozoic aquifer is a fractured (hence inhomogeneous) rather than a porous medium, and the sampling location is relatively close to the tuffcarbonate interface. (4) Temporary perturbations in the ground-water flow regime may have been caused by other detonations in the vicinity.

The data suggest that there is normally no interaction between the equilibrium water table and the cavity. We cannot, however, eliminate the possibility of temporary interactions because of detonation-induced water level fluctuations or of enhanced unsaturated zone recharge through the cavity-chimney system.

In spite of the uncertainties associated with the system, it remains of interest from an RNM standpoint because it is the only documented occurrence of radionuclide contamination of the regionally important carbonate aquifer system.

2.5. Bilby (U3cn)

(Contributors: R. W. Buddemeier, J. Rego, and J. Scholten)

The Bilby Event (located beneath Yucca Flat, Fig. 1-1) was the first detonation below the water table at NTS.²⁹ It was fired on September 9, 1963, had a yield of approximately 200 kt, and produced a cavity with a radius of about 87 m.³⁰ The chimney and cavity began filling with ground water after the chimney collapse, and the water level returned to the pre-shot water table elevation by the end of 1968.²⁹

The geology of the Bilby site is described in Table 2-16; the formations described are found throughout much of NTS with variations primarily in the thickness of rock types. The stratigraphy was determined from lithographic core samples taken during drilling of the satellite well (U3cn5).³⁰ These samples compare well with the lithographic log of the reentry hole (U3cnPS2). Figure 2-7 shows a section through the site and then wells of interest.

Soon after the reentry hole to the cavity (U3cnPS2) was drilled, the pipe was crimped below the water table at 587 m²⁹ by additional chimney collapse. This left the reentry well open for sampling only in the chimney environment, where it was perforated in the interval 512–527 m.

Drilling of the Bilby satellite well (U3cn5) began in September 1965. The well was drilled primarily to determine the depth of the Paleozoic rocks beneath the Bilby site and the distribution and intensity of radioactivity surrounding the emplacement hole. The well, located 122 m SE of U3cnPS2, was drilled to a depth of 922 m, cased to 863 m, and cemented in the interval 837-863 m so that water was produced only from the Paleozoic rocks. At its point of closest approach, the U3cn5 hole is 129 m from the working point and approximately 47 m from the cavity wall. The casing ends 150 m below the working point and approximately 63 m below the bottom of the cavity, which is located only 60 m above the Tertiary-Paleozoic contact. Prior to pump failure in 1981, 3.03×10^6 m³ was pumped from rocks in the Paleozoic carbonate aquifer.³⁰

The U3cnPS2–U3cn5 chimney-satellite pair is considered an ideal location to test for the possibility of Paleozoic aquifer contamination for three reasons: the satellite is cased into the Paleozoic rocks, the event was contained in the overlying tuff near its lower boundary, and rapid transmission of hydraulic pressure during the event suggested the possibility of communication between the two aquifers.³⁰ Tables 2-17 and 2-18 present chronologic summaries of the installation, testing, and sampling of wells U3cnPS2 and U3cn5.

2.5.1. Results

Table 2-19 summarizes the available LLNL and LANL gamma spectrometry and tritium data for samples from well U3cnPS2, and Table 2-20 presents the results for two large-volume samples from U3cn5 analyzed by LLNL. Beetem et al.³¹ studied the dissolved radionuclides in the water samples from the Bilby chimney in 1964. By converting all activities to a common date and unit, the data from 1964 can be compared with the more recent analyses. $T_0 + 14$ y is used as a reference date; it coincides with the first major sampling of U3cnPS2 after the chimney well was filled. The recalculated 1964 data are presented in Table 2-21 along with the results of the two subsequent large-volume samples for comparison purposes. The 1964 data are from a period when the chimney and cavity were still filling with ground water. The water level in the chimney has presumably been in equilibrium with the surrounding water table since 1968.

Garber²⁹ reports some USGS data on ⁹⁰Sr, ¹³⁷Cs, U, gross beta, and gross alpha from pump test samples in the period 1964–66. LLNL submitted aliquots of archived large-volume salt samples to EAL for plutonium and ⁹⁰Sr analyses (see Section 2.1.2) and performed plutonium analysis on one chimney sample. Table 2-22 summarizes the additional data on specific radionuclides from these sources.

2.5.2. Discussion

U3cn5. The satellite well was monitored for more than 10 years without detecting any radioactivity.³³ The two large-volume samples reported in Table 2-20 represent the highest analytical sensitivity achieved, but in neither case was significant activity detected. The tritium levels reported are below the activity of contemporary rainwater at

Layer	Formation	Description	Depth		
1	Alluvium	Medium to coarse sand, gravel and cobble size fragments colored light brown or tan.	0–282 m (0–925 ft)		
2	Timber mountain tuff	Tertiary (younger) volcanic rocks deposited under air, water, or gaseous clouds (welded tuff) colored white to purplish brown.	282–431 m (925–1414 ft)		
3	Indian trail formation	Similar to timber mountain tuff with yellow in upper to reddish-brown in lower regions.	431-860 m (1414-2822 ft)		
4	Lower Paleozoic rocks	Limestone, dolomite, and quartzite.	860-924 m (2822-3032 ft)		

Table 2-16. Bilby stratigraphy.



Figure 2-7. Bilby site cross section.

Table 2-17. Bilby (U3cnPS2) chronology.

Date	Chronology					
9/13/63	Detonation; yield 200 kt; working point at 712.9 m.					
9/23/63	U3cnPS2 drilled into the cavity at a horizontal distance of 10 m from the working point. Total depth 793.4 m; cased to 792.8 m; perforated from 579.1 to 792.5 m.					
10/10/63	Casing collapse; restriction at 587 m.					
7/10/64	Submersible pump lowered to 577 m.					
7/11/64-1/6/65	Pump tests #1 and #2 by USGS. ^a					
6/23/65	Packer set in hole at 561.4 m. Perforations at 512 and 527 m. Submersible pump installed. Intake at 521.8 m.					
7/7-11/65	Pump test #4; rate 54.0 to 57.8 m ³ /day.					
5/3-16/66	Pump test #5; rate 27.2 to 60.0 m ³ /day.					
9/11-14/69	Pumped 186.2 m ³ and collected samples for chemistry and radionuclide analysis.					
6/77	Pump failed.					
9/29/77	New pump lowered to 509 m.					
9/29-30/77	LLNL collected and analyzed a series of samples.					
10/20/80	LLNL collected large-volume sample.					
10/21/81	LLNL collected large-volume sample.					
12/21/81	Pump operable.					
4/18/84	Pump operation questionable.					

^a Pump test samples were analyzed by USGS mobile field laboratory, which determined results from a number of methods: well yield, water temperature, water depth, specific conductivity and pH, alkalinity, dissolved oxygen, and CO₂ content on selected samples.

Date	Chronology					
9/24/65	Drilling started 122 m south from U3cn. Work stoppage delayed completion.					
2/9/66	Total depth 922 m; cased to 863 m.					
3/6/67-9/22/69	Well pumped continuously for 2-1/2 years.					
9/69-5/73	Intermittent pumping. Pump failure after 2.27 \times 10 ⁴ m ³ .					
1979	Pumping resumed.					
10/21/80	Samples taken by LLNL.					
10/20/81	Samples taken by LLNL.					
12/81	Pump failure after 3.03×10^6 m ³ .					

Table 2-18. Bi	lby satellite ((U3cn5)	chronology.
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NTS, and the small ¹³⁷Cs value for the 1980 sample is comparable to values observed for "background" samples from wells far removed from any radionuclide source. It is clear that there was no significant radionuclide migration from the cavity to U3cn5 over the course of the experiment.

U3cnPS2. Before attempting to interpret the changes in chimney water radioactivity, the comparability of the samples and the true precision

and accuracy of the analyses need to be assessed. For the LLNL and LANL analyses over the period 1977–1982, methods were generally consistent and the series of samples taken at closely spaced intervals on 9/29–30/77 can be considered replicates to a first approximation. Based on the ranges of values observed for this suite of samples and the assumption that the large-volume sample is probably more representative than the preceding small

Sample date (m ³	Net vol (L) [salt	Filter					dpm/L (± 1 std de	v %)				Ratio
total)	wt (g)]	used	²² Na	⁴⁰ K	⁶⁰ Co	¹⁰⁶ Ru	¹²⁵ Sb	¹³⁷ Cs	¹⁵⁵ Eu	³ H	²³⁵ U	²³⁸ U	²³⁵ U/ ²³⁸ U
9/27/77 (0.42)	4.1 NA ^a	_	<0.002	48.38 (7.4%)	0.68 (30.00%)	<2.2	1.90 (26.00%)	7.20 (4.90%)	<0.004	$7.77 \times 10^{7^{b}}$	<0.17	59.95 (29.30%)	_
9/29/77 (3.79)	3.8 NA	—	<0.002	45.97 (7.70%)	0.69 (30.00%)	<2.4	2.30 (27.00%)	7.40 (5.10%)	<0.004	$7.93 \times 10^{7^{b}}$	1.80 (13.90%)	47.82 (25.50%)	0.038
9/29/77 (5.68)	3.8 NA	_	<0.002	46.60 (7.80%)	0.66 (30.00%)	<2.4	2.30 (30.00%)	8.30 (4.70%)	<0.004	$8.12 \times 10^{7^{b}}$	1.90 (15.00%)	56.27 (11.90%)	0.034
9/29/77 (8.52)	4.0 NA	—	<0.002	28.36 (13.40%)	<0.17	<1.8	1.50 (29.40%)	5.98 (6.80%)	<0.004	$7.90 \times 10^{7^{b}}$	0.50 (35.60%)	<1.84	
9/29/77 (9.84)	3.9 NA	—	<0.002	57.85 (4.00%)	0.62 (16.00%)	<1.5	2.80 (12.00%)	9.60 (2.50%)	<0.004	$7.59 \times 10^{7^{b}}$	1.90 (8.20%)	58.01 (19.40%)	0.032
9/29/77 (11.35)	4.1 NA	-	<0.002	28.83 (15.10%)	<0.18	<1.8	<0.61	6.60 (6.80%)	<0.004	$7.83 \times 10^{7^{b}}$	<0.17	<1.84	_
9/29/77 (13.25)	3.5 NA		<0.002	32.44 (13.40%)	<0.27	<2.0	1.40 (47.40%)	6.60 (5.80%)	<0.004	$7.73 \times 10^{7^{b}}$	<0.17	<1.84	_
9/30/77 (13.63)	198.0 [85.6]	—	<0.002	35.63 (1.00%)	0.15 (5.20%)	3.90 (3.00%)	2.10 (1.80%)	6.70 (0.90%)	<0.004	$7.73 \times 10^{7^{b}}$	0.60 (3.00%)	14.75 (7.90%)	0.039
10/21/81 NA	227.0 [77.3]	—	<0.002	28.42 (1.60%)	0.03 (15.80%)	<0.71	0.60 (6.90%)	3.60 (17.20%)	<0.004	$8.14 \times 10^{7^{b}}$	0.40 (2.20%)	8.98 (4.10%)	0.045

Table 2-19. Analysis of Bilby chimney well (U3cn) calculated to day 273 (1977).

a NA = not available. b Ref. 32.

Additional data: 9/29/77 ²³⁹Pu 0.19 dpm/mL (6.6%). 9/30/77 ⁹⁵Zr 0.49 (12.6%).

Table 2-20. Analysis of Bilby satellite well (U3cn5) calculated to day 273 (1977).

Sample	Net vol (L) [salt							Ratio					
date	wt (g)]	used	²² Na	⁴⁰ K	⁶⁰ Co	¹⁰⁶ Ru	¹²⁵ Sb	¹³⁷ Cs	¹⁵⁵ Eu	³ H	²³⁵ U	²³⁸ U	²³⁵ U/ ²³⁸ U
10/21/80	1509.0 [593.0]	1.0 μm	<0.002	16.01 (2.20%)	< 0.003	<0.04	<0.01	0.006 (51.30%)	0.20 (44.30%)	19.51	0.10 (15.10%)	1.31 (15.30%)	0.074
10/20/81	715.4 [217.2]	1.0 µm	<0.002	13.35 (1.80%)	<0.003	<0.07	<0.01	<0.003	0.20 (17.20%)	5.16	0.08 (11.60%)	1.09 (16.60%)	0.074

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Table 2-21. U3cnPS2 water activity [dpm/L (\pm 1 std dev %) at T_0 + 14 v].

Nuclide	12/9/64 (Beetem et al.)	9/30/77 (LLNL)	10/21/81 (LLNL)
³ H	2.4×10^8	7.7×10^7 (<1.0%)	8.1×10^7 (<1.0%)
⁶⁰ Co	80	0.15 (5.2%)	0.05 (16%)
¹⁰⁶ Ru	0.8	3.9 (3.0%)	<0.71
¹²⁵ Sb	52	2.1 (1.8%)	0.6 (7%)
¹³⁷ Cs	4.9	6.7 (0.9%)	3.6 (2%)
¹⁴⁴ Ce	1.0 \times 10 ⁻³	<0.1	ND ^a

^a ND = not detectable.

Table 2-22. Miscellaneous analyses of Bilby wells [dpm/L (\pm 1 std dev %) at T_0 + 14 y].

Date (lab)	⁹⁰ Sr	¹³⁷ Cs	²³⁹ Pu
U3cnPS2:			
7/64 (USGS)	<6.4	8.6	a
12/64 (USGS)	—	5.0	
9/77 (LLNL)	_	_	0.019
10/81 (EAL)	1.4	_	0.002
U3cn5:			
10/80 (EAL)	< 0.0085		ND ^b
10/81 (EAL)	ND		ND

^a Not analyzed for.

^b ND = not detectable above background.

samples, we can make two observations based on the data presented in Table 2-19. (1) No significant decrease in tritium content was noted between 1977 and 1981; the apparent 10% increase between the 1981 and 1982 samples is on the margin of significance. (2) The data suggest a 40–80% decrease in the concentrations of ⁶⁰Co, ¹⁰⁶Ru, ¹²⁵Sb, and ¹³⁷Cs between 1977 and 1981, but the range of 1977 values is wide enough so that this must be treated as probable rather than quantitatively certain.

The rather substantial "replicate" variations observed can be ascribed to two possible sources. First, the rubble chimney is undoubtedly highly heterogeneous in the spatial distribution of radioactivity, permeability, and sorption or desorption sites, so that small volumes of water that have been incubated in different regions of the chimney may well have substantially different activities. Second, the inclusion of particulate activity may affect the results. The 1977 samples were unfiltered, while the 1981 sample was passed through a 1.0- μ m filter before evaporation. This was not considered significant at the time, but recent experiments at the U20n chimney well have shown that 20–50% of the total Co, Ru, and Cs activity in a pumped sample can be collected on 0.45 μ m filters (see Sections 2.2 and 3.3). With un-filtered samples, variations in pumping rate, aliquoting, or decanting techniques may cause large variations in the amount of particulate activity included.

In comparing the 1964 data with subsequent analyses (Table 2-21), other considerations must be kept in mind. Beetem et al. filtered their samples through $0.2 - \mu m$ filters, so particulate contamination is less likely than in subsequent samples. However, different methods were employed, and in 1964 the chimney had not completely refilled and was probably still at an elevated temperature. Thus, the geochemical and hydrologic regime may have been quite different from the situation sampled in 1977-82. Taken at face value, the data indicate three conclusions. (1) Tritium levels decreased by 60-70% between 1964 and the 1977-81 period. It is probable that most of this change occurred early in the time interval; Garber²⁹ reports that tritium levels decreased by 14% between July and December 1964, and by an additional 11% between December and the following June. The results are probably more closely related to water table recovery than to steady-state ground-water flow. (2) There is no conclusive evidence of either long-term increase or long-term decrease in the cases of ¹⁰⁶Ru and 137 Cs, but the range of uncertainty is large. (3) ⁶⁰Co decreased in concentration by almost three orders of magnitude, and ¹²⁵Sb concentrations decreased by one to two orders of magnitude.

If we treat tritium as a conservative tracer for water movement and assume that all losses are a result of advection and diffusion, then nuclides that disappear more rapidly must have additional sinks (presumably sorption or precipitation) and those that decrease more slowly must have secondary sources (solution or desorption). Cobalt and antimony clearly fall in the former category and apparently had not equilibrated with the medium by the time of the 1964 sampling. It is tempting to look for evidence for secondary sources in the case of ruthenium and cesium, but it is questionable whether the data are good enough to say with any certainty that their rates of change are significantly different from those of tritium. There is certainly no evidence for any long-term net depletion of the latter two by any means other than water flow.

2.5.3. Hydrology

Garber and Johnston³⁰ report that well logs and core surveys in the interval adjacent to the cavity showed that some but not all of the fractures intersected by U3cn5 contained readily measurable levels of radioactivity. On the basis of physical and mineralogical examination of the fractures, they concluded that both the contaminated and the uncontaminated fractures were preexisting natural features and not shock-induced. Because water levels in U3cnPS2 had not vet recovered to the levels observed in U3cn5 at the time of drilling, the chimney was downgradient from the satellite. They therefore concluded that the radionuclide transport must have been induced by the blast and was not caused by normal ground-water flow. They remarked on, but could not explain, the selective contamination of some but not all of the fractures.

Measurements made following water table recovery show a slight (a few meters of head at the most) apparent gradient from the cavity to U3cn5, but with only two observation points it cannot be determined whether the satellite well is situated directly on the axis of maximum gradient or is off to one side.

Pumping rates at U3cn5 have been modest $(\sim 50 \text{ gpm})$, and the total volume pumped represents no more than 10% of the cavity water content. On a subregional scale, fractured rock systems typically are characterized by nonuniform and indirect preferential flow paths rather than by uniform frontal advance of water. Given the modest natural and imposed gradients and the uncertainties as to the actual direction of preferred flow, it is not clear that we should necessarily expect cavity water to have reached U3cn5 yet. If we compare the Bilby system with the Cambric experiment, we can note that the satellite wells are comparable distances from the cavities, that Cambric is situated in alluvium (hence more likely to yield ground-water flow directly downgradient), that the Cambric satellite has been pumped at much higher rates than U3cn5 (thus inducing a larger gradient), and that breakthrough at the Cambric satellite did not occur until $>10^{6}$ m³ had been pumped from the satellite. This comparison strongly suggests that the negative results at Bilby are inconclusive and supports Claassen's contention that "the test period is too short to conclude that Bilby has not and will not contaminate the Paleozoic aquifer."³⁴

2.5.4. Conclusions

1. Radionuclides from the Bilby cavity did not reach the U3cn5 pumping point over the course of the experiment. This suggests that there has been no rapid or extensive contamination of the Paleozoic aquifer; however, because of the hydrologic uncertainties associated with fracture flow systems and the small gradients and pumped volumes, the results do not rule out the possibility of such contamination.

2. The prompt postshot occurrence of significant activity in natural fractures well beyond the cavity radius indicates that the hydrologic cross section of the radionuclide source considerably exceeds cavity dimensions.

3. Chimney water analyses suggest the existence of an intermediate-term (years to decades) geochemical sink for cobalt and antimony, but not for ruthenium or cesium.

2.6. Bourbon (U7n)

(Contributors: J. Rego and R. W. Buddemeier)

The Bourbon Event (U7n) was detonated on January 20, 1967 and was of low-to-intermediate yield. The working point was at a depth of 560 m in a silty limestone formation.

Satellite well UE7ns was drilled in 1976 to study the geological and geophysical characteristics of emplacement sites that had demonstrated anomalous seismic signals. The well is located 137 m SE of U7n; it was drilled to a total depth of 672 m and cased to 670 m with the bottom 62 m of the casing slotted. At UE7ns, the alluvium-tuff contact is at a depth of 84 m, the tuff-Paleozoic contact is at 503 m, and the static water level is at 601 m. A thrust fault was tentatively identified in the Paleozoic limestone with the top of the fault zone at 594 m.³⁵

A pump was installed in the satellite well, but well yields were so low that only sporadic samples were obtained and it is doubtful that they can be considered fully representative of the formation water. Between 1979 and 1982 tritium values for these samples ranged from 1.0 to 3.7 nCi/L.³⁶ In early 1983, a low-flow solar-powered pump was installed and a large-volume sample was collected for LLNL. Table 2-23 presents the results of the analysis of that sample. Shortly after that collection the system failed and was not reactivated until early 1984. Table 2-24 summarizes the pumping rates and tritium activities reported by REECo since then. Table 2-25 presents the chronology of operations at UE7ns, and Fig. 2-8 shows the vertical cross section of the study site.

2.6.1. Discussion

There are a number of parallels between the Bourbon and Nash sites, but the results obtained to date are significantly different. In both locations, the shot was fired in the limestone just above the water table and the satellite well was completed below the water table within the carbonate. However, Bourbon shows satellite well activities that are only slightly elevated above the regional background for tritium and that give no conclusive evidence for the movement of any other nuclides. The data of Table 2-23 are all at or below the levels attributable to background effects (Section 2.1.1), and there are no indications of the presence of the mobile radionuclides. The only point of interest is the apparent gradual increase in tritium activity since the beginning of the continuous pumping in early 1984 (Table 2-24), but levels are very low compared to those of Nash, and the increase is slow.

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Table 2-23.	Analysis of Bourbon	UE7ns satellite well	calculated to day 175 (1983).
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Sampling date, m ³	Solution vol (L) [salt	Filter				dp	m/L (±	1 std de	v %)				Ratio
pumped	wt (g)]	used	²² Na	⁴⁰ K	⁶⁰ Co	¹⁰⁶ Ru	¹²⁵ Sb	¹³⁷ Cs	¹⁵⁵ Eu	³ H	²³⁵ U	²³⁸ U	²³⁵ U/ ²³⁸ U
6/24/83, 15,675	200.0 [64.4]	0.45 μm	<0.002		0.02 (17.0%)	< 0.05	<0.01	0.09 (6.30%)	<0.004	<3000 ^a	0.007 (78.0%)	<0.27	0.026

^a REECo analysis.

Sample	Volume	³ H	³ H	Rel. % error
date	(gal)	(μCi/mL)	(dpm/L)	(2 std dev)
6/24/83	15,675	$<1.3 \times 10^{-6}$	<2886	<u> </u>
4/23/84	62,300 ^a	1.21×10^{-6}	2686	26.0
5/24/84	102,980	1.98×10^{-6}	4396	15.4
6/15/84	148,010	1.70×10^{-6}	3770	30.9
7/18/84	217,512	2.32×10^{-6}	5150	18.1
8/8/84	260,710	2.51×10^{-6}	5570	12.7
9/5/84	313,600	2.59×10^{-6}	5750	12.3
10/03/84	370,940	2.85×10^{-6}	6327	13.2

Table 2-24. Bourbon REECo tritium log.

^a Pumping restarted after being down since 7/83.

Table 2-25. Bourbon satellite (UE7ns) chronology.

Date	Chronology				
1/20/67	Detonation; hole U7n; intermediate yield; working point 559.7-m depth.				
6/3/76-7/15/76	UE7ns drilled; cased to 670.2 m; casing perforated from 608.2 to 670.2 m. Location 137.1 m from UE7. Water table at 601.1 m, pump at 660 m.				
10/26/78	Tritium reported as 9.5 \times 10 ² dpm/L.				
11/30/78-2/16/82	0/78-2/16/82 Monitored regularly; pump produced small quantities of water.				
2/83	Solar pump installed.				
6/24/83 REECo assisted in sample collection. 200 L processed.					
4/9/84 Solar pump restarted after major parts failure some time after 6/83 sampling.					
4/18/84	Field measurement and large-volume LLNL sampling.				


Figure 2-8. Bourbon site cross section.

The hydrologic setting here is even more complex than in the other well sites. As usual, we lack detailed knowledge of the local natural ground-water gradient, and the very slow pumping rate possible from the satellite well is almost certainly inadequate to induce a significant artificial gradient. In addition, the possible presence of a fault zone between the working point and the satellite well production depth further complicates any interpretation. As at Nash, we are unable to say whether or not the shot cavity has interacted with the local ground-water body.

It is safe to say that there is no indication of significant radionuclide migration from the cavity to the satellite well and that the low water yield makes the site inappropriate for a serious pumping experiment. Further analysis is probably not justified unless routine tritium monitoring indicates the breakthrough of higher activity water or new questions or techniques (e.g., ⁹⁹Tc) are developed.

2.7. Faultless (UC1P2SR)

(Contributors: R. W. Buddemeier, J. Rego, and J. Schweiger)

2.7.1. Introduction

The Faultless site in Hot Creek Valley, central Nevada, is the only site in the RNM Program that is located off NTS. The nuclear event took place in January 1968, at a depth of 975 m in unwelded tuff. The drillback hole (P2SR) was open to 802 m. The water level has been steadily increasing since the event and is currently at 365 m below the surface. Table 2-26 gives the history of activities at UC1P2SR.

As part of an annual sampling program, the Faultless well and two satellite wells (HTH-1, located 915 m SE, and HTH-2, located 762 m SE of UC1P2SR) were sampled during the period of July 18-21, 1983. Field sampling and analysis were carried out by an interagency team consisting of personnel from the USGS, the U.S. Environmental Protection Agency (EPA), and LLNL. The primary purpose of the trip was to evaluate the chemistry, radionuclide content, and water levels in the well system for comparison with previous results. Secondary objectives included intercalibration of the results obtained by the different groups and comparison and assessment of sampling and analytical techniques.

This report contains the results of the LLNL field and laboratory analyses; for comparison and completeness, the USGS field results are also cited. Table 2-27 summarizes depth, time, temperature, dissolved oxygen, Eh, pH, alkalinity, and conductivity data for all field analyses (USGS/EPA as well as LLNL samples). The results are compared with previous Faultless well sample analyses³⁷ and the significance and reliability of the results are discussed. The sampling techniques and the dissolved gas and redox system analyses are discussed in Section 3.2.

2.7.2. Methods

Chemical analyses were carried out by the methods described in Appendix A.

Table 2-26. Faultless (UC1P2SR) chronological history.

Date	Event
1/19/68	Location: Supplemental Central Nevada Test Area. Detonation; hole UC1P2SR; yield range 200–1000 kt working point 975.4 m. Pre-shot water level 170.7 m. Total depth of emplacement hole 1083.3 m.
2/20/68	UC1P2SR location 300 ft north of UC1P2SR. Slant drilled to 1022.6 m. Cased to 851 m. 1022.6 m. Cased to 851 m. Perforations at 350.5 and 851 m.
4/9/68	Water level 653.5 m. Water samples collected on a periodic basis. Sampled from the upper part of the drill hole because of casing constriction at 679.7 m.
9/69-11/70	No sampling. Water level dropped to less than 680 m, below the restriction in the casing.
11/70	Constriction was opened and entire water column sampled at 15-m intervals.
12/2/70	Water level 771 m.
3/31/71	Water level 791 m. Water level rising at about 0.15 m/day.
1972-1981	Well sampled annually by USGS.
11/16/82	Water level 355 m.
7/18-21/83	Joint USGS, EPA, and LLNL sampling effort. Water level 340 m. LLNL samples taken at 545.6, 637.0, 728.5, and 789.4 m.

Depth (ft)	Time (h)	Temp (°C)	DO (ppm)	Eh (mV)	Eh (mV) (NHE)	рН	Alkalinity (ppm CaCO ₃)	Conductivity (ohm-cm) ⁻¹
7/19/83 V	Vell HTH-1	water level	(WL) 534.5 f	it i				
850	1222	24.3	8.0	312		8.13		590
2600	1307		0.9	337		7.73	190	
7/19/83 V	Vell HTH-2	WL 555.7 ft						
504	1000	19.6	10.3	433		7.50	650	
950	1035	24.3	9.0	368		7.88	·····	
7/19/83 V	Vell UC1P2	SR WL 1116	ft					
1150	1500	22.9	7.6	250	227	9.43		225
1190	1549	24.1	8.0	253	248	9.43		200
1290	1620	24.7	10.4	264	259	9.63		210
7/20/83								
390	0936	24.6	10.4	257	263	9.63		200
490	1116	24.3	10.7	319	319	9.62		215
590	1134	25.1	10.0	363	331	9.61		225
690	1230	24.9	8.6	256	250	9.61	112	225
790	1311	25.1	11.3	261	267	9.63		215
790	1334		8.2	306	293	9.71	99	
890	1431	25.0	10.9	296	286	9.62	100	215
990	1500	25.8		308	303	9.32		240
1090	1531	26.3	9.3	318	319	9.29		240
1090	1608		8.2	340	319	9.30	144	
7/21/83								
190	1627	25.4	10.5	242	269	9.10		320
290	1208	26.3	10.5	439	293	9.03		350
390	1253	26.4	11.6	281	281	9.10	124	350
2390	1305		7.2	228	262	9.10		
2490	1415	26.3	9.1	243	268	9.10		340
2590	1430	26.4	9.3	268	268	9.01		300
2590	1530		7.3	268	268	9.12		

Table 2-27. Faultless field measurements.

For the radionuclide analysis, a weighed aliquot of each sample was analyzed for tritium by liquid scintillation counting (LSC), using procedures that have been shown to yield satisfactory agreement with REECo analyses of UE2ce (RNM-2S) samples. The deepest Faultless sample (2590 ft) was also given a long LSC count on a Beckman 9800 in the spectrum search mode, and the spectrum above the tritium region was compared with a background spectrum in an effort to find and identify any alpha or nontritium beta activity. A 500-lambda aliquot of each sample was flamed onto a stainless steel planchet and counted on an alpha spectrometry system designed and calibrated for the measurement of low levels of neptunium and plutonium. The filters through which the laboratory samples were passed were also used to filter the water retained from the field analyses (total combined volume filtered ~ 6 L) and were analyzed for gamma emitters using Ge(Li) detectors and the GAMANAL spectral analysis program. A 1-L Marinelli beaker containing filtered water from the 2590 ft sample was also given a long count on a low-background, high-efficiency Ge(Li) system.

2.7.3. Results

Radiochemical. Tritium activities are summarized in Table 2-28.

The results of gamma analyses of all sample filters and the UC1P2SR 2590 ft filtrate are summarized in Table 2-29. In general, no significant gamma activity was detected; as noted, the 2590 ft filter appeared to contain ¹²⁵Sb. We view these results with suspicion and consider sample or filter contamination in the laboratory to be the most plausible explanation.

Direct alpha spectrometry of small filtered sample aliquots detected no plutonium (<0.005 pCi/mL) or neptunium (<0.01 pCi/mL).

A long-count LSC spectral search on the 2590 ft filtrate revealed no alpha activity or beta activity with energy greater than that of tritium (detection limit \sim 0.5 pCi/mL).

Table 2-28. Faultless sample tritium activities.

Sample	Depth (ft)	μ Ci/mL (±2 std dev			
HTH-1	2600	$<1.0 \times 10^{-6}$			
HTH-2	950	$< 1.0 \times 10^{-6}$			
UC1	1790	$< 1.0 \times 10^{-6}$			
UC1	2090	1.49×10^{-5} (4.4%)			
UC1	2390	$2.04 \times 10^{-2} (0.5\%)$			
UC1	2590	2.07×10^{-2} (0.2%)			

Table 2-29.Upper limits of Faultless samplegamma activity.(All activities are in pCi/mL.)

Nuclide	Particulate $>0.45\mu^{a}$	UC-1 2590 ft filtrate ^b
⁷ Be	$< 8.0 \times 10^{-4}$	
⁴⁰ K	$< 1.1 \times 10^{-3}$	
⁵⁴ Mn	$<$ 5.3 \times 10 ⁻⁵	$<$ 5.0 \times 10 ⁻⁴
⁶⁰ Co	$<$ 5.3 $ imes$ 10 $^{-5}$	$<$ 4.9 \times 10 ⁻⁴
¹⁰⁶ Ru	$<$ 4.4 $ imes$ 10 $^{-4}$	$<$ 5.1 $ imes$ 10 $^{-3}$
¹²⁵ Sb	$< 1.2 \times 10^{-4^{\circ}}$	$< 1.6 \times 10^{-3}$
¹³⁷ Cs	$< 9.4 \times 10^{-4}$	$<$ 5.6 \times 10 ⁻⁴
¹⁴⁴ Ce	$< 3.4 \times 10^{-4}$	
²³⁸ U	$<$ 2.7 \times 10 ⁻³	
²⁴¹ Am	$<$ 3.5 $ imes$ 10 $^{-4}$	

 $^{\rm a}$ Except as noted below, all sample filters had nuclide upper limit values < blank filter values; reported values are for blanks.

^b No identifiable peaks were observed; limits are based on 3 \times background std dev.

 c UC1P2SR 2590 ft filter showed 4.0 \times 10 $^{-4}$ (±22%) pCi/mL $^{125}Sb.$ The value is suspect.

Chemical. Table 2-30 presents both field measurement results on the LLNL samples and the corresponding laboratory measurements on the pressurized samples returned to the laboratory. In addition to the dissolved oxygen in ppm, the ratio of the observed value to the saturation value at the temperature and altitude of the measurement is also reported. Measurements were made on unfiltered samples except for the laboratory alkalinity and conductivity measurements, which were made after filtering through a prefilter and a 0.45- μ m Nuclepore filter. The laboratory alkalinity values are averages of one or more Hach titrator determinations and one or more pH titrations; there was no substantial or systematic difference between the results of the two methods. Sulfide was not detectable (<0.1 ppm) in either field or laboratory-analyzed samples.

Table 2-31 gives the results of cation determination by ICP expressed as mg/L. Table 2-32 presents the results of anion analyses.

2.7.4. Discussion

The only radionuclide reliably detected at a significant level was tritium. The values and vertical distribution of ³H were similar to the data previously reported by the USGS; however, the concentrations observed are modestly but significantly higher (10–15%; decay corrections were not applied and would make the differences even

	HTH-1 HTH-2			UC1P2SR				
	2600 ft	950 ft	1790 ft	2090 ft	2390 ft	2590 ft		
Field samples ^a						· ·		
рН	7.73	7.88	9.71	9.30	9.10	9.01		
Eh mV	337	368	293	319	262	268		
DO ppm	0.84	8.4	7.6	7.6	6.7	6.8		
DO/DO sat.	0.13	1.26	1.14	1.17	1.03	1.05		
Alk. ppm CaCO ₃	190	_	99	144	124	_		
S =	ND ^b	ND	ND	ND	ND	ND		
Lab samples								
pH ^a	7.75	7.90	9.09	8.86	8.75	8.91		
Eh mV ^a	235	356	319	305	264	255		
DO ppm ^a	0.9	17.5	9.1	6.9	7.6	9.0		
DO/DO sat. ^a	0.11	2.08	1.08	0.83	0.90	1.08		
Alk. ppm CaCO ₃ ^c	196 ± 5	$148~\pm~10$	87 ± 2	106 ± 5	118 ± 6	112 ± 11		
Cond. mho/cm ^c	570	330	230	290	355	320		
S =	ND	ND	ND	ND	ND	ND		

Table 2-30. Faultless sample field/laboratory measurements.

^a Unfiltered.

^b ND: $S^{=} < 0.1$ ppm.

^c Filtered, 0.45 μ m.

	Detection						
Element	limit	HTH-1	HTH-2	F-1790	F-2090	F-2390	F-2590
Aluminum	0.008	0.028	0.029	0.029	0.100	0.059	0.051
Boron	0.004	0.010	1.83	0.110	0.162	0.244	0.236
Cadmium	0.002	0.004	< 0.002	< 0.002	< 0.002	< 0.002	< 0.002
Cobalt	0.003	0.004	< 0.003	< 0.003	< 0.003	< 0.003	< 0.003
Copper	0.002	0.031	0.012	0.011	0.005	0.029	0.003
Iron	0.002	0.013	0.314	0.044	0.088	0.175	0.872
Lithium	0.001	0.013	0.152	0.046	0.055	0.065	0.060
Manganese	0.0003	0.040	0.047	0.007	0.013	0.058	0.020
Molybdenum	0.004	0.019	0.101	0.074	0.138	0.043	0.040
Nickel	0.008	0.023	0.009	< 0.008	< 0.008	< 0.008	< 0.008
Phosphorus	0.042	0.142	0.139	0.074	0.124	0.139	0.112
Lead	0.016	0.041	0.028	0.016	0.018	0.017	0.024
Selenium	0.044	0.053	0.044	< 0.044	0.045	0.064	0.069
Silicon	0.004	13.3	27.8	6.59	9.68	11.6	11.4
Strontium	0.012	0.543	0.027	0.246	0.203	0.139	0.120
Uranium	0.068	0.044	< 0.00	< 0.068	< 0.068	< 0.068	< 0.068
Vanadium	0.003	0.005	0.007	< 0.003	0.007	0.024	0.019
Zinc	0.001	0.110	0.109	0.045	0.027	0.257	0.073
Zirconium	0.002	< 0.002	0.015	< 0.002	< 0.002	< 0.002	0.002
Calcium	0.004	37.4	2.78	1.94	2.48	2.52	2.21
Potassium	0.26	2.02	1.49	1.42	1.10	1.06	0.935
Magnesium	0.0004	5.41	0.161	0.099	0.090	0.084	0.083
Sodium	0.004	19.2	139	48.1	63.2	72.4	68.6

Table 2-31. Cation (ICP) analysis of Faultless samples made in the period of July 18-20, 1983.

Notes: Ignore all values less than the detection limit.

The detection limit is 4 times the standard deviation. Results are in micrograms per milliliter (or ppm).

Table 2-32.	Anion analyses of	Faultless samples.	(All values are in mg/L.)
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					UC1	P2SR	
Ion	Method ^a	HTH-1	HTH-2	1790 ft	2090 ft	2390 ft	2590 ft
F -	IC	9.8	ND ^b	1.0	1.3	1.7	1.6
Cl ~	IC	21.0	3.2	4.9	5.5	7.4	6.9
NO_3^-	IC	ND	ND	ND	ND	Trace	ND
SO₄	Ι	38.6	3.4	20.5	27.4	33.5	32.6
Alkc	AA	201	146	87	108	119	119

^a IC = ion chromatography; uncertainties $< \pm 1.5\%$. AA = autoanalyzer.

^b Not detectable.

^c Alk is carbonate alkalinity (HCO₃⁻ + CO₃⁼) expressed as mg/L CaCO₃, \pm 3 mg/L.

larger) than the 1981 USGS results.³⁷ It remains to be seen whether this is a systematic interlaboratory difference or whether the tritium values have actually increased. If the increase is real, it has interesting implications in terms of the relationship of the sampling points to the source and the movement of tritiated water (HTO).

Although the counting methods used were relatively insensitive compared with sample concentration or radiochemical separation techniques, no credible evidence of other radioactivity was found. The small ¹²⁵Sb activity indicated for one of the filters is suspect. If the apparent increase in

³H activity proves to be real, it may be desirable at some time in the future to perform more sensitive analyses for some of the mobile fission products.

The laboratory alkalinity determinations (Hach titrator, pH titration, and autoanalyzer) are in close agreement and consistent with earlier USGS profiles, as are the conductivity data.³⁷ Comparison of filtered and unfiltered alkalinity data suggests that the two shallower UC1P2SR samples may have contained some particulate carbonate or other titratable base (max ~40 ppm at 2090 ft). Agreement between LLNL cation and anion results and the 1981 USGS data is reasonable;

in general, it appears that the analytical results obtained by the two laboratories are satisfactorily comparable.

Except for the possible shift in tritium activity discussed above, the only other significant change from the earlier results was an increase in pH (relative to the 1982 USGS results) of 0.2–0.4 in the depth interval below 2100 ft. Temperatures were consistent with 1982 data, although lower than in earlier samplings.

Depth profiles of the measured parameters remained qualitatively consistent with earlier observations, with different but internally consistent regions above 1900 ft and below 2100 ft and transitional values in between. This may relate to the drilling report of loss of circulation at 1979 ft, and it suggests different hydrologic regimes above and below this depth.

The data, in conjunction with USGS data from earlier samples, suggest that the upper and lower zones have distinctly different chemical signatures that are reasonably consistent over time and depth. It is interesting to note that HTH-1 and HTH-2 have chemical characteristics that are distinctly different both from each other and from both of the zones observed in UC1P2SR. Prior to 1982, there was a marked thermal gradient in the lower region of UC1P2SR (USGS data previously distributed); this appears to have disappeared rather abruptly. Figure 2-9 shows the history of observed temperatures at 2590 ft.

In addition to the hydrologic implications of the four different geochemical regimes sampled in the three wells, three interesting points are raised by the secular changes observed in UC1P2SR:

1. The abrupt change in thermal gradient near the bottom (see Fig. 2-9) is not characteristic of a static cooling curve and probably represents



Figure 2-9. Observed temperatures at 2590 ft at UC1P2SR (Faultless site).

the combined effects of the heat source and the ground-water flow regime.

2. The shift in pH in the lower zone is hard to evaluate. Since the pH increase was noted between 1982 and 1983 and the temperature dropped prior to 1982, temperature effects on the chemical equilibria cannot readily account for the shift unless the rate of equilibration is very slow.

3. If the apparent increase in tritium concentrations is real, it suggests migration of higher activity water toward the bottom of the well and indicates that it might be useful to sample for other radionuclides.

In view of the data record and the observed changes in the well water characteristics, it seems appropriate to initiate a review and interpretation of the results obtained to date before resampling this system.

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3. Methodological Investigations

3.1. The Phantom Europium Migration

(Contributors: R. C. Evans, K. V. Marsh, R. W. Buddemeier, and J. Rego)

¹⁵⁵Eu is a reasonably abundant fission product (fission yield \sim 0.03%) with a 4.7-y half-life. It is known to occur as a persistent component of detonation cavity and surface contamination activity at the NTS. Gamma spectral analysis of large-volume evaporated water samples from two satellite wells (Nash and Bilby) consistently yielded indications of low levels (tenths to hundredths of dpm/L) of ¹⁵⁵Eu. In the case of the Nash satellite, other radionuclides were also observed (see Section 2.4). This suggestion of europium mobility was further supported by the results of an unrelated study of radionuclides in the soil column below an abandoned disposal site at LLNL, where tritium and ¹⁵⁵Eu appeared to occur at penetration depths greater than those observed for other nuclides.¹ Because there were no literature reports of or obvious geochemical reasons for such environmental europium mobility, we set out to verify or disprove its presence by three concurrent approaches: (1) recounts of several archived samples that had initially been measured nearly one ¹⁵⁵Eu half-life ago; (2) a careful review of possible interferences at the ¹⁵⁵Eu gamma-ray energies and the calculational procedures used by the GAMANAL computer code to identify and quantify ¹⁵⁵Eu, and (3) redissolution of an NTS groundwater salt sample (Nash satellite well, April 18, 1984) that had yielded an apparent ¹⁵⁵Eu value by bulk gamma spectroscopy, followed by radiochemical separation for europium and measurement of the yield-corrected activity of the purified sample.

1. The recounted samples yielded apparent activities comparable to the initial determinations. Although these results were not absolutely conclusive because of relatively large statistical uncertainties and some variation in the peak ratios of the gamma rays, they did not show the decrease to be expected from ¹⁵⁵Eu decay.

2. The calculational library used for our GAMANAL runs identifies ¹⁵⁵Eu from two or three of the following gamma energies: 105.3 keV (22.8%), 86.6 keV (32.2%), and 60.01 keV (1.32%). At the very low apparent counting rates observed, the 60-keV peak is not above background, so 86.6

and 105.3 are the primary energies used for ¹⁵⁵Eu measurement. Both of these are subject to interferences. The region around the 86.6-keV peak is generally complicated by lead x rays generated in the detector shield, and by lead and bismuth x rays from the natural decay series elements in the sample. The 105.3-keV peak overlaps the 105-keV thorium x ray. To calculate the disintegration rate of a nuclide, GAMANAL utilizes a leastsquares weighted average of as many nuclear gamma rays as possible. X rays are not included in the calculations because their absolute intensities are not constant; thus, they are "left over" and may be mistaken for gamma rays from other nuclides if they are similar in energy. Consequently, the 105-keV x rays from thorium could be interpreted as resulting from a gamma ray from ¹⁵⁵Eu. A similar argument applies to the 86.6-keV peak, and the presence of both peaks in a spectrum may satisfy the GAMANAL requirements for detection of ¹⁵⁵Eu, at least with a relatively large error and low probability. We examined the spectra and compared the ratios of the various europium and thorium peaks, but could find no consistently convincing evidence to prove or disprove the presence of ¹⁵⁵Eu.

3. The salt analyzed was an 88.8-g aliquot of the April 18, 1984 Nash satellite sample, which originally showed an apparent ¹⁵⁵Eu concentration of 0.38 dpm/L by bulk gamma count. The sample was dissolved in dilute HCl, and a known amount of stable europium carrier was added. EuF₃ was precipitated with HF and redissolved with boric acid and nitric acid. The hydroxide was then precipitated with NH₄OH, washed, and redissolved in 8M nitric acid. This solution was passed through a preconditioned Dowex AG1-X8 column (50-100 mesh), then eluted with 8M nitric acid. This step elutes the europium, but leaves thorium on the column. After this separation, the europium was again precipitated as the hydroxide, washed, redissolved in HCl, and finally reprecipitated as the oxalate. After washing and drying, the europium oxalate was ignited and weighed as Eu₂O₃ to determine yield and the oxide sample was gamma counted. Separation of the thorium as well as radium and its daughters eliminated the major possible interferences. Because of the greatly improved counting geometry and reduced total sample activity, a much lower detection limit for ¹⁵⁵Eu was achieved.

The purified sample, when recounted on our low-background Ge(Li) system, yielded no discernable peaks indicative of either thorium or europium. The chemical europium yield was 33%. Based on this and the detector calibration and background data, we calculate that the ¹⁵⁵Eu concentration in the original water sample must have been less than 0.0035 dpm/L. Compared with an original bulk salt determination of 0.38 dpm/L, this result indicates that essentially all of the apparent ¹⁵⁵Eu activity was in fact caused by interfering photons, probably dominated by the thorium x ray.

It should be noted also that all of the samples that showed apparent europium activities were either soils or ground waters that had probably interacted with the Paleozoic carbonate aquifer at NTS. Although thorium analyses are not available, it is possible that these samples might have higher natural thorium concentrations than the groundwater samples from tuffaceous and alluvial aquifers, which did not show apparent europium activities. We conclude that direct gamma spectrometry measurements of low levels of ¹⁵⁵Eu in environmental samples are unreliable unless a confirming radiochemical separation is performed, all three of the europium gammas are observed with appropriate intensity ratios, or a half-life measurement is performed.

3.2. Pressurized Sample Integrity

(Contributors: R. W. Buddemeier, J. Rego, and J. Schweiger)

During July 1983, a joint sampling effort was conducted at the Faultless Event site (Central Nevada Test Area) by the USGS, EPA, and LLNL personnel. UC1P2SR and two satellite wells, HTH-1 and HTH-2, were sampled; the results of the chemical and radiochemical analyses are reported in Section 2.7. Field and laboratory observations caused us to review and investigate the performance of the pressurized "thief" samplers used for down-hole sampling in wells not equipped with pumps. The results are reported below.

3.2.1. Sampling

Two similar types of down-hole samplers were used, both made of stainless steel with Oring sealed couplings. The USGS 6-L sampler, which featured an electromechanically activated valve, was used to take top and bottom samples in the satellite wells and a vertical profile in the Faultless well. The sampler was closed at the surface, attached to the valve-actuating mechanism on the end of the cable, lowered to the desired depth, and then opened and closed to collect the sample. The samplers used by LLNL consisted of two 1.5-L USGS barrels coupled together with a similar valve head at the upper end. The bottom of both barrels and the top of the lower one were fitted with manual valves. In use, the sampler was sent down after evacuation with the two middle valves open and the top (electromechanical) and bottom valves closed. This sampler was used on a second run at preselected depths (based on previous chemical and radiochemical data) after the USGS sample was taken. For the LLNL samples, the sampler was evacuated at the surface to a pressure of 0.1-0.2 Torr using a Welsh Duo-Seal pump, closed, and then opened and reclosed at the desired sampling depth. Prior to use, the evacuated sampler was left connected to the vacuum manifold for 30-60 min with the pump valved off to check for leaks; no measurable pressure increase was observed. All samplers were lowered and activated with the calibrated cable on the EPA field van.

The USGS sampler was opened after retrieval and decanted into a large stainless steel container from which aliquots were taken for the various field and subsequent laboratory analyses. The internal valves on the two separate component barrels of the LLNL sampling assembly were closed before the barrels were decoupled, and one sealed barrel from each sample was shipped back to LLNL for analysis. The second barrel of each pair was sampled in the field by attaching a hose barb fitting to one end and slowly opening top and bottom valves to allow the water to flow out. Efforts were made to minimize exposure to the atmosphere of those sample aliquots intended for redox-sensitive measurements by avoiding the first and last samples, keeping the drain tube full and the flow rate slow, etc. However, field observations indicated that most of the samples taken with the evacuated samplers were pressurized and effervescent on decanting, and showed levels of dissolved oxygen (DO) comparable to the samples taken without evacuation of the sampler.

3.2.2. Laboratory Procedures

At LLNL, the sealed, pressurized sample barrels were drained in essentially the same fashion as in the field; however, because the field samples were effervescent and showed oxygen supersaturation, a system was developed to sample the pressurized gas from the barrels before sampling

the water. A vacuum-tight hose barb fitting was attached to the upper end of the barrel, and to that was attached a four-way connector with a vacuum gauge, a mass spectrometry gas sample tube, and a valved connection to a laboratory vacuum pump. This system was pumped down, valved off from the pump, and checked for leaks. The upper sampler barrel valve was opened. Pressure in the attached manifold was recorded, and the gas sample tube was closed, removed, and submitted for analysis. The water samples were then decanted and promptly analyzed for O₂, Eh, pH, and sulfide to investigate the effects of storage in the pressurized barrels; Eh and dissolved oxygen were followed as a function of time after decanting in one of the samples.

3.2.3. Results

Analytical results are tabulated in detail in Section 2.7. As mentioned above, the field samples were generally effervescent, and with the exception of HTH-1, gave dissolved oxygen readings in excess of the local saturation value (see Table 2-30). On the basis of rather crude pressure-volume estimates, the four UC1P2SR samples all released 10–15 cc of gas when vented into the evacuated sample tubes. The results of mass spectrometric analysis of these gas samples (on a water free volume % basis) are given in Table 3-1, which also includes the standard composition of dry air for comparison.

The laboratory samples were also effervescent (again with the exception of HTH-1) when decanted, and showed values at or near oxygen saturation (Table 2-30). After the initial measurements of oxygen and Eh, the HTH-2 lab sample was shaken vigorously and remeasured; DO decreased from the original 17.5 to 11.4 ppm, and Eh dropped from 356 to 155 mV. The same sample was then purged for 2 min with argon gas, after which the DO dropped to 3.7 ppm and the Eh to 117 mV.

These results indicate that, at atmospheric pressure, all but one of the samples taken with the

Table 3-1. Composition (vol%) of gas from pressurized samples of UC1P2SR.

Gas	1790 ft	2090 ft	2390 ft	2590 ft	Air
N ₂	84.09	81.26	85.17	85.46	78.09
$\overline{O_2}$	14.82	17.72	13.78	13.51	20.95
Ār	1.068	0.974	1.03	1.004	0.93
CO ₂	0.020	0.039	0.02	0.023	0.03

"evacuated" samplers was supersaturated with a gas having a composition closely approximating that of air. The fact that the oxygen content of the gas is reduced relative to that of air may suggest that some of the introduced oxygen reacted with the sample; if this was the case, it would imply that the original sample contained chemical species that were reduced relative to equilibrium with the atmosphere. The correlation of DO with Eh described in the preceding paragraph suggests that observed Eh values may be affected by the oxygen gas dissolved in the water.

3.2.4. Discussion

When an air-filled sampler is opened at a depth significantly below the water surface, the hydrostatic pressure forces all or most of the gas into solution. When the containing pressure is released at the surface, the solution is supersaturated and effervesces. Because there is no credible mechanism for producing ground waters naturally supersaturated with nitrogen and argon, we are forced to conclude that the gases observed were contaminants carried down with the sampler and injected in situ. There are two possible mechanisms by which this may have happened: (1) The leak test used in the field did not detach the samplers from the evacuated manifold, so it did not verify that the upper valve was leak-free at atmospheric pressure, and (2) an air bubble may have been trapped in the electromechanical valveactuating head on the cable.

Based on the volumes of gas collected, the measured concentrations, and the assumption that the HTH-1 sample represents uncontaminated ground-water conditions, each of the samples except HTH-1 must have contained a minimum (since we have no estimate of gas volume lost by effervescence between depressurization and DO measurement) of ≥ 100 cc of air. In a 3-L sampler, this corresponds to a pressure on the order of 0.05 atm (~40 Torr).

Although the data presented in Section 2.7 (other than those for DO and Eh) are unlikey to be affected by this contamination, the chemical implications of this exposure are significant in terms of the potential uses of these samplers. The data of Table 3-1 suggest that something like 25% of the oxygen in the original air may have reacted with the sample or sampler; this would correspond to about 5 cc, or 0.2 mmol. This is ample to destroy any redox-sensitive equilibria in the system, because all of the reducible species are present at less than millimolar concentrations (see Table 2-23). In addition to affecting any direct

measurements of redox speciation, this sample alteration could potentially affect the distribution of trace elements between dissolved and particulate phases in cases where different oxidation states have radically different solubilities or sorption characteristics.

The effect of the carbon dioxide in the included air can also be estimated; the amount available would be a few μ mol. Because these samples contained bicarbonate/carbonate concentrations at the mmol level, they would be relatively well buffered and micromoles of carbon dioxide would probably have a negligible effect on either pH or carbonate system concentrations. This would not necessarily always be the case, however; if a poorly buffered water were sampled with an air-filled sampler, pH perturbations would be possible. The data presented in Table 3-1 and Section 2.7 also suggest that no significant loss of CO₂ occurred by degassing or gas purging, because the evolved gas CO₂ concentrations did not differ significantly from those of air. The pH of the Faultless samples was high enough so that essentially no undissociated aqueous carbon dioxide or carbonic acid should have been present.

Dissolved oxygen and sulfide can both be measured reliably at the 0.1-ppm level. For these measurements to be significant, the sample would have to contain considerably less than 1 cc/L of introduced air, which implies the ability to evacuate and maintain the entire sampler assembly at a final pressure of less than 0.001 atm with reasonable confidence.

3.3. Filtration Studies

(Contributors: R. W. Buddemeier and J. Rego)

The question of appropriate filtration procedures for RNM ground-water samples is a vexing one. Noted below are four of the issues that need to be considered:

1. Intercomparison of samples. Most geochemists report analytical data for samples that have been filtered through 0.45- or 0.2- μ m filters. There is no standardized procedure for environmental radioactivity studies, and a review of past analyses of RNM well samples shows that various practices have been followed.

2. Interpretation of results. The nuclides of greatest interest from the standpoint of migration potential are those present in dissolved form; inclusion of particulate activity in this category results in an overestimate of the activity available for migration. On the other hand, small colloidal

particles may be transported by ground-water flow, and any strongly sorbed species associated with such particles would therefore be more mobile than models based solely on solution chemistry would indicate.

3. Equilibrium. If the sample collected is for any reason not fully equilibrated between dissolved and sorbed fractions, or if chemical changes occur between sampling and analysis that affect the solution-solid equilibrium, the activities measured will be a function of time and any chemical changes in the sample may not represent the true *in situ* conditions.

4. Sample integrity. Field filtration is tedious and increases the likelihood of sample contamination, while laboratory filtration may undersample the particulate fraction (because of settling in the storage container) and, as noted above, may yield results representing postsampling processes rather than the original conditions.

LLNL large-volume samples have generally been either unfiltered or filtered through a $1.0-\mu$ m cartridge filter (see Section 2.1.1). The results to date have not yielded evidence of any significant difference between these two methods. During 1983 and 1984, we experimented with various filtration methods. Our initial efforts made use of 0.45- μ m Nuclepore filters, but results were unsatisfactory. In addition to operational inconvenience caused by the use of low-capacity filters, we consistently observed high blank values for unused filters; in most cases involving satellite well samples, the blank was considerably higher in activity than the actual sample filters.

A LANL study of the Nuclepore filters has shown that they contain measurable amounts of a number of radionuclides, and that at least some of this activity is washed out into the filtrate at time of use.² This makes them particularly inappropriate for use with low-level samples.

We have subsequently switched to bag-type filters (Fin-L-Filters, a hydrophilic terpolymer formulation supplied by Cole-Parmer), which are used in a standard cartridge-type filter holder. Available in 0.45- and 0.2- μ m nominal sizes, as well as a felt prefilter, they have low activity blanks (see below) and a higher capacity and throughput than the Nuclepores. Although initial results seem satisfactory, we are continuing our evaluation.

In an effort to assess the amount of activity associated with very small ($<0.2-\mu$ m) particles, we have initated a series of studies using a commercially available (Millipore) ultrafiltration system. This system passes the feed solution across a series of very fine membrane filters under pressure; the filtrate is collected and the flow system recycles the retentate into the feed stock. The retentate (feed) solution may be reduced to a volume limited only by the volume of the flow system by use of a continuous-flow recycling process. Membrane filter cartridges are available with nominal ratings of 100,000, 10,000, and 1,000 molecular weight units (MWU). To date, we have used the 100,000 MWU membrane, which has an equivalent pore size rating of approximately $0.006 \,\mu$ m.

Two experiments have been performed, both using water samples from the Cheshire chimney well (U20n—see Section 2.2 for a discussion of the Cheshire system). This water has radionuclide levels high enough to provide good analytical sensitivity for investigations of the distribution between different fractions. The first sample analyzed was a composite sample consisting of 2-L samples taken at corrected flowmeter readings of 81,370, 96,660, 107,125, 120,425, and 130,480 gal and a 19-L sample taken at a corrected flowmeter reading of 71,603 gal. All of these samples were field filtered through the same multiple 0.45- μ m filter assembly. After a 1-L aliqout of the 19-L sample was gamma counted in a Marinelli beaker, the samples were combined to yield a single 28-L composite, which was then further processed through the Millipore ultrafiltration unit rated at approximately $0.006 \,\mu$ m. The retentate (~140 mL) was evaporated to dryness and gamma counted, and the filtrate was recounted in the Marinelli beaker system. Table 3-2 presents the results, expressed in units of dpm/L of composite solution, for the gamma analyses of these samples.

We also analyzed by our standard laboratory procedures a 200-L sample taken at a corrected flowmeter reading of 131,860 gal. This sample was filtered through a prefilter and $0.45-\mu m$ filter in the laboratory and then evaporated to dryness. The resulting salt sample was homogenized and gamma counted (see Section 2.2). Table 3-3 summarizes the total, filtrate, and particulate (>0.45- μm) activities expressed as dpm/L.

Because the small volumes analyzed for the composite sample made the analyses relatively insensitive for the less abundant nuclides, a second

Nuclide	Filters (0.45 μm)	Ultrafilter retentate	Ultrafiltrate	Total
²² Na	0.92 (31%)		2.86 (26%)	3.8
⁴⁰ K	—	0.84 (33%)	-	0.8
⁵⁴ Mn	0.87 (16%)	<0.01	<0.9	~0.9
⁶⁰ Co	7.27 (3%)	0.05 (27%)	<0.9	~7.3
¹⁰⁶ Ru	218 (1%)	3.62 (9%)	<21.3	~222
¹²⁵ Sb	79.1 (2%)	17.2 (2%)	1709 (4%)	1805
¹³⁴ Cs	1.87 (8%)	0.07 (31%)	2.44 (22%)	4.4
¹³⁷ Cs	3555 (1%)	95.5 (5%)	3596 (2%)	7151
¹⁴⁴ Ce	46.8 (5%)	1.27 (15%)	_	~48.1
¹⁵² Eu	23.2 (2%)	0.58 (10%)	—	~23.8
¹⁵⁴ Eu	30.1 (2%)	0.73 (6%)	-	~30.8
¹⁵⁵ Eu	53.9 (6%)	2.06 (5%)	_	~56.0

Table 3-2. Analyses of U20n 28-L composite sample calculated to day 253.5 (1983). Units are dpm/L (\pm 1 std dev %).

and more extensive experiment was subsequently designed using a 188-L sample of U20n water (collected at 56,330 gal). Figure 3-1 shows the experimental arrangement; the water was passed through a complete series of bag filters before passing over the ultrafiltration unit, and an electric drum stirrer was used to mix the water in the barrel in order to keep the particulates suspended. Ultrafiltration was continued until the retentate fraction was reduced to approximately 3 L; this was transferred, the barrel was spray-rinsed with about 4 L of deionized water, and the rinse fraction was also ultrafiltered and combined with the sample. At the end of the process, the ultrafilter was back-flushed into the retentate fraction, and the filtrate and retentate fractions were dried and gamma counted, as were all filters. Table 3-4 presents the activities measured for each fraction, normalized to dpm/L. The assembly was thoroughly cleaned, fresh filters were installed, and the entire process was repeated with a "blank" of 200 L of laboratory deionized water. Samples of filtrate and retentate from both the U20n sample and the deionized water were submitted for ICP analysis. In addition to the filters used in the blank experiment with deionized water, a set of unused filters was also counted.

Table 3-5 presents the results of the ICP analysis of both the original solutions and the

Tabl	e 3-3.	U20n	200-L	sam	ıple	cal	cula	ted	l to
day	253.5	(1983).	Units	are	dpn	ı/L	(±	1	std
dev	%).								

Nuclide	Filters + prefilter (0.45 μm)	Filtrate	Total
²² Na	0.23 (38%)	2.67 (9%)	2.90
⁴⁰ K	3.1 (12%)	8.45 (7%)	11.6
⁵⁴ Mn	0.2 (32%)	0.38 (33%)	0.58
⁶⁰ Co	1.7 (5%)	3.32 (2%)	5.0
¹⁰⁶ Ru	35.2 (5%)	156.9 (2%)	192
¹²⁵ Sb	37.5 (4%)	2067 (1%)	2105
¹³⁴ Cs	0. 9 (4%)	3.83 (6%)	4.7
¹³⁷ Cs	1341 (1%)	5135 (1%)	6476
¹⁴⁴ Ce	6.8 (11%)	25.8 (7%)	32.6
¹⁵² Eu	3.5 (3%)	14.5 (3%)	17.8
¹⁵⁴ Eu	4.8 (3%)	18.9 (3%)	23.7
¹⁵⁵ Eu	8.5 (3%)	36.2 (3%)	44.7



Figure 3-1. Schematic of ultrafiltration assembly.

Nuclide	Prefilter filter bag	0.45-μm filter bag	0.20-µm filter bag	0.006-µm membrane	Retentate	Filtrate	Total
²² Na	NL ^a	NL	NL	NL	<0.10	2.11 (3.8%)	~2.1
⁴⁰ K	, <0.10	<0.09	<0.09	<0.06	3.71 (19.4%)	2.22 (18.9%)	~5.9
⁵⁴ Mn	0.28 (8.5%)	0.04 (27%)	0.03 (31%)	<0.01	0.21 (57%)	<0.03	~0.56
⁶⁰ Co	2.61 (0.9%)	0.37 (2.0%)	0.32 (1.9%)	0.04 (11%)	1.23 (3%)	<0.01	4.57
¹⁰⁶ Ru	30.77 (2.0%)	13.64 (2.5%)	10.81 (2.8%)	1.41 (19%)	93.99 (3.2%)	13.40 (26.4%)	164.0
¹²⁵ Sb	12.91 (2.2%)	5.25 (1.3%)	3.78 (1.3%)	0.64 (4%)	68.62 (2.6%)	1671.3 (1.1%)	1763
¹³⁴ Cs	0.21 (20.5%)	0.08 (9.0%)	0.06 (15%)	NL	1.23 (9.3%)	2.24 (3.6%)	~3.8
¹³⁷ Cs	500.9 (0.8%)	141.2 (1.0%)	95.21 (0.8%)	31.11 (0.8%)	1931 (1.1%)	3276 (0.8%)	5975
¹⁴⁴ Ce	7.34 (6.4%)	1.93 (9.6%)	1.43 (11%)	0.38 (20%)	15.42 (9.2%)	NL	~26.5
¹⁵² Eu	4.01 (1.7%)	1.01 (2.6%)	0.75 (2.0%)	0.14 (19%)	8.51 (2.8%)	<0.07	~14.4
¹⁵⁴ Eu	5.61 (1.2%)	1.42 (1.7%)	1.05 (1.4%)	0.19 (10.8%)	11.54 (2.0%)	<0.16	~20.4
¹⁵⁵ Eu	9.34 (1.7%)	2.45 (3.2%)	1.77 (2.9%)	0.43 (3.1%)	16.38 (5.4%)	<0.34	~30.4

Table 3-4. U20n 188-L ultrafiltered samples calculated to day 253.5 (1983). Units are dpm/L (\pm 1 std dev %).

^a No limit calculated.

Notes: There is an interfering gamma ray for ²²Na and ¹⁵⁴Eu at 1274 keV. Calculated zero time for measured activities is day 253.5 (1983). Zero time calculated for limits is count time.

Table 3-5. Ultrafilt	ation-study r	retentate and	alysis (µg	z/mL).
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Element	U20n 9/83	U20n retentate concentration 1:70	DI water	DI retentate concentration 1:70
Aluminum	2.13	24.8	<0.020	<0.020
Boron	0.106	1.58	< 0.004	<0.004
Copper	< 0.003	0.006		0.161
Iron	0.294	3.26	0.005	0.019
Lithium	0.161	0.286	0.001	0.023
Manganese	0.108	0.449	< 0.0005	0.012
Molybdenum	0.021	0.109		0.003
Nickel	< 0.008	0.008	< 0.008	0.010
Phosphorus	< 0.124	0.141	_	0.080
Lead	0.026	0.142	_	0.060
Silicon	33	122	0.090	0.041
Strontium	0.004	0.068	< 0.012	0.028
Titanium	-	0.103	_	0.002
Uranium	< 0.084			0.078
Zinc	< 0.008	0.065	_	0.275
Zirconium		0.018	< 0.002	0.001
Calcium	1.58	8.28	<0.004	0.478
Potassium	3.11	24.7	<0.536	0.504
Magnesium	0.165	0.456	0.024	0.173
Sodium	73.05	176	0.041	0.208

deionized water blank and the retentate fractions after these samples were ultrafiltered. The filtrate:retentate volume ratio in both cases was approximately 70. Most elements show some concentration in the retentate, with factors ranging from less than 2 to 17. As reported in Section 2, the total salt concentration in the retentate was approximately 15 times the ultrafiltrate salt concentration. This suggests that approximately 18% of the total salt content <0.45- μ m was present as colloidal particles in the 0.006- to 0.45- μ m size range.

Table 3-6 presents the results of all blank determinations, both on unused filters and on filters and retentate from the deionized water experiment (for the prefilter bag, results are presented from a specimen used in processing a low-activity Nash satellite sample, which has essentially zero activity compared with the Cheshire water). The only significant activity noted (discounting the ¹⁵⁵Eu value for the 0.45- μ m blank bag, see Section 3.1) is ¹³⁷Cs in the deionized (DI) water experiment. As may be seen from Tables 3-2 through 3-4, ¹³⁷Cs shows by far the highest activity found in both the particulate and dissolved fractions of the Cheshire sample; we believe that these "blank" values represent the level of cross-contamination

to be expected from reuse of the same filtration system, even with decontamination efforts between uses. Table 3-6 shows total dpm per sample; when the values of Table 3-4 are converted to total dpm, it can be seen that the ¹³⁷Cs carryover is $\leq 0.01\%$. Table 3-4 also shows that small but measurable levels of activity (~1%) were retained on the ultrafilter even after back-flushing.

As discussed in Section 2.2, the $0.45-\mu m$ filtrate activities, the ultrafiltrate activities, and the total sample activity inventories are all in reasonable agreement, particularly when realistic experimental uncertainties are considered in place of the tabulated values based on counting statistics only. From the standpoint of the filtration experiment, there are some differences in activity distribution that may be significant (see Table 3-7). The composite sample retentate showed appreciably lower activity levels than the 188-L sample retentate, the >0.45- μ m fraction of the composite sample was considerably more active than the same fraction for the large-volume samples, and the 188-L sample showed less ¹³⁷Cs and ¹²⁵Sb in the >0.45- μ m fraction than did the 200-L sample.

These differences may be related to the fact that the filters had different amounts of water passed through them—28 L in the case of the

Table 3-6. Ultrafiltration-study blanks (total dpm). Calculated zero time is count time (August-October 1984).

Nuclide	Prefilter filter bag ^a	0.45-µm blank bag	0.20-µm blank bag	0.006-µm blank membrane	DI 0.20-µm 200-L bag ^b	DI 0.006-µm 200-L membrane	DI retentate (200 L)
⁷ Be	<7.1	NL ^c	<6.3	<3.9	NL	<3.9	NL
²² Na	NL	<1.2	<0.8	<0.4	<1.0	<0.5	< 0.4
⁴⁰ K	<25.7	<25.8	<17.7	<12.4	<21.4	<12.4	<15.7
⁵⁴ Mn	<0.9	<1.1	<0.8	<0.4	<1.0	<0.4	< 0.3
⁶⁰ Co	<1.2	<1.2	<1.0	<0.5	<0.9	<0.5	<0.3
⁶⁵ Zn	<2.0	NL	<1.8	<1.1	NL	<1.0	NL
Nb/ ⁹⁵ Zr	<0.9	<2.1	<0.9	<0.80	<1.7	<0.80	<0.6
¹⁰³ Ru	<0.9	NL	<0.7	<0.49	NL	<0.40	NL
¹⁰⁶ Ru	<8.0	<9.8	<7.2	<4.3	<8.5	<4.2	<3.0
¹²⁵ Sb	<2.4	<3.0	<2.0	<1.4	<2.6	<1.3	<0.9
¹³⁴ Cs	NL	NL	NL	NL	NL	NL	NL
¹³⁷ Cs	<1.0	<1.3	<0.9	<0.55	4.4	61.53	10.37
					(24.9%)	(1.9%)	(5.2%)
¹⁴¹ Ce	<1.1	NL	<0.9	<0.95	NL	<0.5	NL
¹⁴⁴ Ce	<4.8	<5.3	<3.9	<4.4	<4.4	<2.3	<2.4
¹⁵² Eu	NL	<2.0	<2.2	<1.6	<1.7	<1.3	<1.0
¹⁵⁴ Eu	NL	<3.5	<1.0	<1.3	<3.0	<0.7	<1.1
¹⁵⁵ Eu	NL	4.7 (27.3%)	<1.8	<2.2	<1.8	<1.2	<1.2

^a Prefilter is from 8/83 Nash sample at 200 L.

^b Followup filtration set after U20n - 200 L of DI water.

^c No limit calculated.

Nuclide	0.45-μm 11/83 (adj.) ^a	0.45-µm 8/84	0.006 retentate 11/83 (adj.)	0.006 retentate 8/84
⁷ Be	<273	<3.5	<10.6	NL ^b
²² Na	27.9 (7.8%)	<3.5 NL	48 (7.8%)	<18.7
⁴⁰ K	627.5 (11.6%)	<17.1	168.6 (33%)	697.7 (19.4%)
⁵⁴ Mn	28.8 (21.1%)	7.3 (27%)	<2.7	39.1 (57%)
⁶⁵ Zn	<6.8	<1.6	<5.7	NL
⁶⁰ Co	248.9 (2.1%)	70.3 (2.0%)	9.8 (26.8%)	230.8 (3%)
Nb/Zr	<3.4	<0.9	<3.9	<14
¹⁰³ Ru	<25	<3.3	<7.0	NL
¹⁰⁶ Ru (622 keV)	5980 (2.6%)	2564 (2.5%)	724 (8.5%)	1.767×10^4 (3.2%)
¹²⁵ Sb	2423 (3.0%)	987.9 (1.3%)	3430 (1.7%)	1.29×10^4 (2.6%)
¹³⁴ Cs	162.9 (3.9%)	14.4 (9.0%)	14 (30.5%)	230.9 (9.3%)
¹³⁷ Cs	2.52×10^5 (0.9%)	2.654×10^4 (1.0%)	1.92×10^4 (4.6%)	3.63×10^5 (1.1%)
¹⁴¹ Ce	<24.4	<3.4	<6.5	NL
¹⁴⁴ Ce	1.16×10^3 (10.6%)	363.2 (9.6%)	254 (15%)	2.90×10^{3} (9.2%)
¹⁵² Eu	614.6 (2.5%)	189.9 (2.6%)	117.5 (9.2%)	1.60×10^{3} (2.8%)
¹⁵⁴ Eu ^c	858.2 (2.2%)	267 (1.7%)	145 (6.1%)	2.17×10^{3} (2.0%)
¹⁵⁵ Eu	1445 (2.9%)	460.2 (3.2%)	412 (4.5%)	3.08×10^3 (5.4%)

Table 3-7. Comparison of filter sets for the U20n ultrafiltration study. Units are total dpm (\pm 1 std dev %). The limits are calculated at count time. No half-life corrections are calculated.

^a 11/83 activities have been multiplied by 188/28 to compare with 8/84 activities on an equal volume basis.

^b No limit calculated.

^{c 154}Eu has an interfering gamma ray for the calculation of ²²Na.

composite sample, 200 L for the "standard treatment" large-volume sample, and several times that amount (because the ultrafiltration recycling was passed through the entire filter string) for the 188-L sample. This may have resulted in disaggregation of the larger particles initially trapped on the filters, with some of the activity transferred to the smaller size fractions. This must remain speculative, because there were other differences that could have affected the results-the samples were stored for different lengths of time between collection and processing, and the prefilter and filter combinations used were not identical for any of the samples. A second experiment is planned using multiple filters at each stage and single-pass filtration prior to the ultrafiltration process in order to resolve some of these questions.

The most significant result, however, is that essentially none of the manganese, cobalt, cesium, nor europium, and very little of the ruthenium activity passes through the ultrafilter, even though significant amounts of these nuclides are detected in the 0.45- μ m filtrate. This strongly suggests that much of the transition element and lanthanide activity that is treated as dissolved on the basis of traditional methods is in fact associated with particles with dimensions >6 nm. If correct, this observation will have profound implications for both geochemical and transport modeling of these elements. Related to this is the observation that the retentate/filtrate activity ratios in Table 3-4 are much higher than the chemical ratios in Table 3-5. Although chemical analyses were not performed for most of the elements present as radionuclides,

the trends in the data suggest that "dissolved" radioisotopes may not act as tracers for their "dissolved" stable counterparts.

In spite of remaining uncertainties, four conclusions can be reached:

1. Significant amounts of activity (relative to "dissolved" concentrations) may be found associated with suspended particles, and the particulate/ dissolved activity ratios are different for different nuclides.

2. There is at least some evidence for the presence of activity on particles that are too small

to be filtered on conventional methods, especially in the case of transition and lanthanide elements (see Section 2.2).

3. The type and technique of filtration employed are clearly capable of producing alterations in the relative amounts of activity assigned to the various particle size ranges and the dissolved phase.

4. Further work on these issues is needed.

3.4. References

1. Stone, R., M. R. Ruggieri, L. L. Rogers, D. O. Emerson, and R. W. Buddemeier, 1982, *Potential for Saturated Groundwater System Contamination at the Lawrence Livermore National Laboratory*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-53426.

2. Thompson, J., 1984, Los Alamos National Laboratory, Los Alamos, NM, personal communication, 1984.

(Contributors: M. R. Ruggieri and R. W. Buddemeier)

LLNL and DRI have initiated a collaborative study of water and radionuclide transport processes in the unique environment provided by the steady-state recharge plume beneath the ditch that carries the effluent water from the RNM-2S pumping experiment. This environment contains a complete range of conditions from natural desert floor through a continuum of moisture contents and unsaturated flow conditions to saturated or nearly saturated flow directly under the ditch. The geologic medium is tuffaceous alluvium, which is characteristic of much of NTS, and the input water has been well characterized over time in terms of its radionuclide content, chemistry, and flow rate. Because the plume is located in the shallow subsurface and may reasonably be treated as a steady-state structure, experimental access is simple and inexpensive, and reproducible and progressive experimental designs can be employed. We expect the results to provide valuable insights into unsaturated zone water transport processes and various aspects of radionuclide transport under both saturated and unsaturated conditions. Plume studies will provide information directly relevant to the potential of ditch recharge to alter the tritium signal at RNM-2S. Unsaturated transport results will provide an important information base relevant to evaluation of the potential for ground-water contamination by enhanced local recharge through craters and at other point sources of potential contamination (e.g., decontamination washdown areas). Finally, access to a shallow saturated alluvial flow regime will offer the potential for performing inexpensive and technically feasible field-scale tracer experiments that will be applicable to models of solute behavior in the deeper aquifers.

DRI is investigating the water budget and chemical and stable isotopic characteristics of the ditch and discharge pond and is modeling the recharge plume; these efforts will be reported elsewhere. This section deals primarily with soil moisture and tritium results obtained in the joint LLNL-DRI sampling effort.

In 1982, DRI submitted to LLNL for soil tritium analyses 7 core samples from a 20-ft borehole located 3 ft from the edge of the ditch. The water obtained by freeze-drying showed tritium activities of about 1230 dpm/mL at a depth of 2 ft, and 4200–5400 dpm/mL over the interval 4 –20 ft. Three of these samples were subse-

quently baked out under vacuum at 120°C; the second (bound water) fractions recovered had tritium activities identical to the freeze-dry (loose water) fractions, indicating isotopic equilibrium across a range of soil pore sizes.

These preliminary analyses demonstrated the existence of unsaturated zone tritium activities comparable to that of contemporaneous ditch water (RNM-2S effluent). In early 1983, DRI cored samples at locations 50 and 100 ft from the ditch in order to determine background values and the outer boundary of the plume. The results of tritium analyses on these soil samples are reported in Table 4-1.

Based on these preliminary studies, an experimental plot was designed and installed during the summer of 1983. Figure 4-1 shows the location of the study site relative to RNM-1, RNM-2S, and the ditch. Also indicated on the figure are the locations of three flumes installed by DRI to measure water flow, and sampling stations established in the outflow pond to study seasonal changes in recharge water characteristics (to be reported elsewhere).

Figure 4-2 shows the detailed layout of the instrument plot. The initial design called for instrumentation to a depth of 20 ft, but the soil auger used to drill the holes could not consistently penetrate a gravel layer encountered in the 12- to 15-ft range. Instrumentation includes suction lysimeters for soil water sampling, tensiometers, resistance cells, and a neutron probe access tube to monitor soil moisture content. Drive core samples were taken for tritium analysis at selected depths during the installation process. Figure 4-3 is a vertical section across the study site showing all of the core and instrument positions in profile.

Since the time of installation, the instruments have been monitored weekly and lysimeter samples (when available) have been collected monthly by DRI field personnel.

Table 4-1. Camplic alea sull n activit	e 4-1. Cambric area soil ³ l	H activit [,]
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Distance from	Sample	³ H			
ditch (ft)	depth (ft)	dpm/mL	pCi/mL		
50	0-2	$125.2~\pm~24.4$	56.4 ± 11		
50	2-4	5.3 ± 2.2	2.4 ± 1		
50	8-10	0.4 ± 0.4	0.2 ± 0.2		
50	18-20	$0.1~\pm~0.1$	$0.06~\pm~0.6$		
100	0-2	11.0 ± 2.2	4.94 ± 1		
100	8-10	<0.1	< 0.06		
100	18-20	<0.1	< 0.06		



Figure 4-1. Cambric site ditch and pond.



Figure 4-2. Cambric unsaturated zone study site.

4.1. Results

Based on measured flow rates and estimates of maximum evapotranspiration rates, DRI personnel have estimated that infiltration losses through the bottom of the ditch amount to 175 gal/min between the first and third flumes. Assuming a 1-km flume separation, 2-m ditch width, and a soil porosity of 25%, this yields a piston flow pore velocity of 20 cm/d or 73 m/y. Since the pumping experiment has been running for more than 9 y and the water table is at a depth of about 220 m, it seems likely that even with allowances for dispersion the recharge plume has reached the water table.¹

Although a few of the FY84 samples remain to be analyzed, the tritium results from a majority of the samples collected are reported in Table 4-2 and Fig. 4-4. Counting uncertainties are generally less than 1%. For better comparison with the ditch



Figure 4-3. Vertical view of soil instruments and samples at the Cambric study site.

water activity, data reported by LANL²⁻⁴ are reported along with the more sporadic LLNL analyses. There appears to be a consistent 5-10% difference between LLNL and LANL ³H analyses of the ditch water, the cause of which is under investigation. In the discussion that follows, the lysimeter data are compared with the LLNL ditch values for internal consistency. The 3-ft lysimeters generally failed to yield samples because of dry soil conditions; some of the other lysimeters failed (loss of vacuum) even though soil moisture should have been adequate to obtain samples. The soil core and initial lysimeter sample activities are portrayed schematically in Fig. 4-5. The soil moisture data are presented in Appendix B. A detailed interpretation of these results has not yet been completed; it will be reported later. A qualitative summary of the major features of the results is shown by the moisture zones indicated in Fig. 4-5. Although none of the instruments indicates fully saturated conditions, very wet soils are observed next to the ditch and extending out at least 5 ft at a depth of 12-15 ft. Elevated moisture

levels are found at the 11-ft installation, but at 21 ft the soils are dry (presumably the normal desert condition). Seasonal variations and transient rainfall responses are observed in the shallower and drier locations, but these have little or no effect on the near-ditch plume.

The basic pattern of tritium activities is quite consistent. Table 4-2 shows that lysimeter A-6 is very close to equilibrium with the ditch water, with time lags of no more than a few weeks. Lysimeter C-12 appears to lag the ditch water in activity by about 1.5 y. Lysimeter D-6 has recently shown a significant increase in activity; if this represents the transmission of the the tritium breakthrough to this sampling point, a transit time of about 7 y is implied. E-12, which did not function for nearly a year, has recently started yielding samples that appear to be in equilibrium with D-6. This would indicate primarily horizontal rather than vertical migration of the water in this zone.

The results demonstrate the feasibility of using soil sampling techniques to intercept and identify various phases of the RNM-2S tritium



Figure 4-4. Chronological record of ditch and lysimeter ³H values.

Table 4-2.	Cambric dite	L-1	L-2	A-6	C-12	D-6	E-1
Date	Ditch	L-1	L-2	A-6	<u> </u>	D-6	E-1
83131	4375 ^b						
145	4356 ^b						
154	4294 ^b						
154	4455						
160	4296 ^b						
168	4264 ^b						
173	4268 ^b						
181	4238 ^b						
188	4215 ^b						
195	4161 ^b						
202	4183 ^b						
215	4113 ^b						
223	4093 ^b						
229	4196 ^b						
237	4249 ^b						
245	4210 ^b	`		4050	-010		
250	h			4258	5010		
251	4232 ^b						
255	b			4205	5047	354	
258	4064 ^b						
262	h				5151	495	
265	4051 ^b						
271	4020 ^b						
271	4186			4268		374	
272				4206	5029		
283					5181		
287	4026 ^c						
292	_			4395			
83300	3965°						
306	4028 ^c						
311	_			4090	4842		
314	4021 ^c						
318				3971			
320	4019 ^c						
325				4002	4739	443	
326	3895°						
332		2270	1294	3967	4858	338	
335	3857 ^c						
339		2267	1391	3955	4849		
343	3854 ^c						
348	3857						
356	3812 ^c						
363	3784 ^c						
84003		3000		3953	4669		
012	3760 ^c						
018	3742 ^c						
026	3764 ^c						
030				3771	4613		
033	3768 ^c						
040	3769 ^c						
047	3732 ^c						
054	3712 ^c						
061	3710 ^c						
065		2929	1993	3694		568	
068	3685 ^d		,				
076	3552 ^d						
082	3574 ^d						
089	3552 ^d						
096	3574 ^d						
103	3508 ^d						
111	3441 ^d						
84114				3881			
117	3463 ^d						
	0 100						

Table 4-2. Cambric ditch and lysimeter ³H (dpm/mL).^a

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Table 4-2. (Continued)

Date	Ditch	L-1	L-2	A-6	C-12	D-6	E-12
121		<u> </u>	t	3832	4596	672	
124	3441 ^d						
132	3463 ^d						
139	3463 ^d	•					
146	3441 ^d						
153	3419 ^d						
156	3863 ^d			3898	3667	871	
159	3397 ^d						
166	3330 ^d						
200	3330 ^d						
212	3759			4032		871	853
220	3756						
221	3754						
230	3241 ^d						
248				3437	4282		
263	3197 ^d						
275				3443	4231	1154	

^a See Figs. 4-1 and 4-2 for instrument depths and locations.

^b LANL data, Ref. 2. ^c LANL data, Ref. 3.

^d LANL data, Ref. 4.



Figure 4-5. Soil core, lysimeter, and ditch ³H activities (dpm/mL).

front migrating through the soil column, and to use activities to determine transport rates. In addition to continued monitoring of these installations, we plan to slant-drill some holes into the presumably saturated zone beneath the ditch to determine stratigraphy and vertical flow rates, better define the shape of the plume to assist in calculating the rate and extent of recharge to the water table, and investigate the possibility of performing active field tracer experiments.

4.2. References

1. Jacobson, R., 1984, Desert Research Institute, Reno, NV, personal communication.

2. Daniels, W. R., and J. L. Thompson, 1984, Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1982 to September 30, 1983, Los Alamos National Laboratory, Los Alamos, NM, LANL Progress Report LA-10121-PR.

3. Thompson, J. L., 1984, "T Analysis of RNM-2S Water Samples," Los Alamos National Laboratory, Los Alamos, NM, LANL memorandum RNM-INC11-4/84-1 to RNM distribution.

4. Thompson, J. L., 1984, Desert Research Institute, Reno, NV, personal communication.

5. Geochemical Modeling Activities

(Contributors: D. Isherwood and J. Rard)

5.1. Introduction

Ruthenium and technetium data bases were compiled for the EQ3/6 geochemical codes to study the potential for ruthenium and technetium migration from a nuclear explosion site. The ruthenium data base compiled for EO3/6 was abstracted from Rard's 1985 critical review of ruthenium thermodynamic data.¹ The 476 references listed by Rard contain essentially all of the thermodynamic data for ruthenium that were available in the literature at the time the document was written. The values recommended by Rard were used to create the data files for both solid phases and aqueous species. In all, 9 ruthenium solids and 26 aqueous species were included in the data base. The gases and liquids were omitted from the main EQ3/6 data base as they exist only at temperatures and conditions that are well outside the range of values for geochemical applications involving aqueous solutions. The technetium data base was created in a similar way using the 1983 compilation of technetium thermodynamic data by Rard.² Thermodynamic data for 8 aqueous species and 15 solids were used to create the technetium data base. Copies of the ruthenium and technetium data files and a more detailed discussion of the modeling studies can be found in Ref. 3, which was authored by Isherwood. Wolery^{4,5} describes the EQ3/6 code package, which is made up of two codes, EQ3NR and EQ6.

5.2. The Cambric Site—A Field Study

At the site of the Cambric nuclear test at NTS, a field study of radionuclide migration has been ongoing since 1975. The Cambric device was detonated at a depth of 294 m in tuffaceous alluvium in May 1965. In 1974, two wells were drilled—one into the Cambric cavity region and a satellite well (RNM-2S) 91 m to the south. The objective of the study was to determine the potential for radionuclide migration from an underground nuclear test.

Pumping of the satellite well began in October 1975 and has continued since that time. No radioactivity was observed for the first two years. With further pumping, tritium concentrations began to increase in samples from the satellite well and to decrease in the cavity.⁶ Attempts were made to measure radionuclides other than tritium using large-volume samples that were evaporated in the field using a distillation apparatus. ¹⁰⁶Ru and several other radionuclides were observed in the samples by Coles and Ramspott.⁷ The ¹⁰⁶Ru/ ³H ratios were nearly the same for all water samples from both wells. Coles and Ramspott suggest that ruthenium and tritium traveled at the same rate from the Cambric cavity to the satellite well. Their finding contradicts the prediction, based on laboratory sorption measurements, that ¹⁰⁶Ru would migrate at a slower rate than tritium.

5.3. Geochemical Modeling of the Cambric Site

The ruthenium data base was used with the EQ3NR geochemical code to evaluate the ruthenium geochemistry at the Cambric site. EQ3NR calculates from water analyses the distribution of ions, ion-pairs, and complexes and determines whether the water sample is saturated with various minerals.⁴ For this study, water analyses of samples taken from the satellite well (RNM-2S) were used as input to the code (see Table 5-1). Ruthenium was added as a component. Because the oxidation potential of the system is unknown, the system was assumed to be oxidizing (Eh = + 0.8 V) based on the work of Winograd and Robertson.⁸ They report that dissolved oxygen has been observed in a variety of aquifers in the south-central Great Basin, Nevada, and in all valley-fill aquifers in the southern Arizona portion of the Basin and Range Province.

Under oxidizing conditions, where the solid phase controlling solubility is RuO₂, the calculated solubility limit is 2.1×10^{-11} M. For comparison, a study of the effect of reducing conditions (Eh = -0.1 V) was also made. Under reducing conditions, the solubility (1.5×10^{-23} M) is controlled by Ru(c). The pH also strongly controls ruthenium solubility (Fig. 5-1). For example, in oxidizing conditions, as the pH is lowered, the solubility decreases to a lower limit of 10^{-18} M at pH = 6.3. Below that pH value, the solubility again increases and the dominant stable aqueous species becomes Ru(OH)₂⁺⁺ instead of RuO₄⁻.

Cambric site (RNM-2S).	
_	RNM-2S ²
Component	(mg/L)
Na ⁺	61
K ⁺	9.9
Ca ^{+ +}	18
Mg ^{+ +}	5.5
A1 ^{+ + +}	NA ^b
Fe(total)	NA
SiO ₂	66
HCO ₃	170
Cl ⁻	15
SO4	40
F -	0.5
NO ₃	NA
HPO4	NA
рН	8.1
O ₂	NA

Table 5-1. Ground-water analyses for the

^a Values shown are averages of analyses for samples taken from January to December 1982 provided by L. Benson (USGS).

^b NA = net available.

How does the solubility limit calculated for the Cambric ground water compare with the ruthenium concentration in the cavity prior to pumping? To make this comparison, we must first make two assumptions: (1) the chemical composition of ground water in the satellite well represents the composition of ground water in the cavity well, and (2) the pH of the water in the cavity at approximately the depth of the perforations in the satellite well is also the same (i.e., 8.3).*

To calculate the total ruthenium content at the Cambric site, we must know the total of all ruthenium isotopes generated at the time of the nuclear detonation (Table 5-2). Based on the ¹⁰⁶Ru found in a sample collected from the bottom of the cavity (8.3 dpm/mL, which is equivalent to $3.4 \times 10^{-16} M$ at 15 y after detonation[†]) and the percent of total ruthenium represented by the ¹⁰⁶Ru isotope (0.0007%) also at T = 15 y, the concentration of ruthenium in solution is $4.5 \times 10^{-11} M$. The EQ3NR code predicts that ruthe-



Figure 5-1. Plot drawn from calculations using successive runs of EQ3NR. Both the speciation and solubility of ruthenium are dependent on pH.

nium solubility in an oxidizing environment where RuO_2 in the stable solid phase is 2.1 × 10^{-11} *M*. Given the uncertainties in the data base and the assumptions regarding the composition and pH of the cavity water, the two numbers are remarkably close.

The EQ3NR code predictions coincide with the field observations. The calculated ruthenium solubility matches the observed values when corrected for the presence of other ruthenium isotopes. The dominant aqueous ion, RuO_4^- , is negatively charged and is therefore likely to migrate. Clues to the reasons for the apparent discrepancy between the laboratory and field measurements can be found in the Eh-pH diagrams.

5.4. Ruthenium Eh-pH Diagrams

To test the effect of oxidation potential or pH on speciation and solubility over a wide range of Eh-pH conditions, an Eh-pH diagram for the aqueous species of ruthenium was generated using a version of the code SOLUPLOT, a code designed to calculate and plot complex Eh-pH diagrams for systems with up to 35 aqueous species and solids containing a particular element.¹⁰

^{*} A second sample from the bottom of the cavity had pH = 11, but was apparently contaminated with drilling fluid (high calcium and lithium values were reported) and was not used in this evaluation.

[†] Fifteen years was chosen for comparison with the data reported by Coles and Ramspott.⁷

Isotope ^b	Percent	Half-life	Atoms at $T = 0$	Atoms at $T = 15$ y
¹⁰¹ Ru	6.75	Stable	8.1 × 10 ²¹	8.1×10^{21}
¹⁰² Ru	6.65	Stable	8.0 \times 10 ²¹	$8.0~\times~10^{21}$
¹⁰⁴ Ru	5.86	Stable	7.0×10^{21}	7.0×10^{21}
¹⁰⁶ Ru	4.64	357 d	5.6 \times 10 ²¹	1.7×10^{17}

Table 5-2. Ruthenium isotopes generated at the Cambric Event.^a

^a Yield = 0.75 kt.

^b Isotope percentages are from Fleming.⁹ Because of their short half-lives, ruthenium isotopes 103, 105, 107, and 108 are not included in the calculations.

Figure 5-2 shows that the stability fields for the dominant aqueous ions are highly dependent on the concentration of ruthenium in solution as well as the Eh-pH of the system. The boundaries between positively and negatively charged species shift with changes in concentration. If the experimental conditions were at concentrations or pH's different from those in the field, then one of the positively charged species shown in the diagrams could be dominant and sorption onto the solid would occur. The most likely species are $Ru(OH)_2^{++}$ and $Ru_4(OH)_{12}^{4+}$.

As concentrations decrease, the stability field of the negatively charged ion RuO_4^- increases. This may explain the migration of ruthenium, because the ruthenium becomes more dilute (and therefore more negatively charged) as water moves into and away from the cavity (assuming that negative ions migrate and positively charged ions are likely sorbed onto the rock). Kinetics could also affect the results of laboratory sorption experiments using ruthenium. For example, if the conversion of the positively charged to negatively charged species is slow relative to the time of the laboratory experiments, the decrease in concentration of the ruthenium remaining in solution as the positively charged ruthenium species is sorbed onto the solid phase would not favor the formation of RuO_4^- . In this case, the laboratory conditions would not be representative of field conditions.

The interpretation of the results of the geochemical modeling relies heavily on theory and the reliability of the data base. Ruthenium experiments that combined leaching and sorption on crushed alluvium gave distribution coefficients $(K_d's)$ of 1000 to 3400 mL/g.¹¹ Given the low solubility of ruthenium in the Cambric ground water, the $K_d's$ could be a measure of precipitation rather than sorption. In a reducing environment (Eh = -0.1 V), the solubility of ruthenium in the +3 oxidation state (i.e., $1.5 \times 10^{-23} M$) is much less than in oxidizing conditions. The formation of positively charged ions could also be a contributing or a major factor. Given the complexity of ruthenium chemistry, the geochemical models used in this study provide new answers, but not the only answers.

Like any model, the Eh-pH diagrams and the predicted solubilities and speciation using EQ3NR are based on the assumption that the total system is at equilibrium, that is, all couples shown on the Eh-pH diagram or considered in the EQ3NR calculations are in equilibrium with each other. Because of their dynamic nature, many natural systems do not achieve equilibrium. Models are also dependent on the number of species in the thermodynamic data base and on the quality of the data. Models are best used as an interpretive tool in combination with field and laboratory experience.

5.5. Technetium Geochemistry

Technetium solubilities were also calculated for the Cambric ground water using EQ3NR. In oxidizing conditions, the stable aqueous species is the pertechnetate ion, TcO_4^- , and solubility is virtually unlimited. In a reducing environment (Eh = -0.1 V), the calculated solubility is 1.8 \times $10^{-14} M$ using Tc₃O₄ as the solubility constraint and $3.0 \times 10^{-8} M$ using amorphous TcO₂·2H₂O. Based on the thermodynamic values compiled by Rard,² EQ3NR predicts that the relatively insoluble Tc_3O_4 is the most stable oxide for a reduced form of technetium. However, laboratory studies of technetium have consistently given solubilities of approximately 10^{-5} to 10^{-8} M in what were believed to be anoxic conditions.^{12,13} This suggests that a thermodynamically less stable form of technetium, possibly the amorphous $TcO_2 \cdot 2H_2O_1$ controls technetium solubility-at least under laboratory conditions.

An Eh-pH diagram was created using the SOLUPLOT code to predict the dominant aqueous species of technetium present within the stability field of water [Fig. 5-3(a)]. Other aqueous species may be present in lesser amounts, but are



Figure 5-2. Eh-pH diagrams generated for aqueous species of ruthenium by SOLUPLOT. Solid phases are not shown.



Figure 5-3. Eh-pH diagrams generated for technetium by SOLUPLOT. (a) Stability fields are shown only for the aqueous species of technetium. (b) Stability fields shown are for both aqueous and solid species at $[T_c] = 2.0 \times 10^{-7} M$, the MPC_w for ⁹⁹Tc.

not shown on the Eh-pH diagram. In Fig. 5-3(b), the stability fields for both the aqueous and solid phases are indicated. The boundary between the stable aqueous phases (e.g., TcO_4^-) and the solid phases is drawn where the concentration of technetium in solution (2.0 \times 10⁻⁷ M) equals the maximum concentration of ⁹⁹Tc in drinking water as permitted by the Federal Government (10CFR60 App. B). The TcS₂ and Tc₂S₇ stability fields shown in this diagram are a function of the amount of sulfate or sulfide present in the Cambric ground water. The Eh-pH diagram shown in Fig. 5-3(a) predicts that at equilibrium the dominant aqueous species of technetium will be TcO₄⁻ in an oxidizing to moderately reducing environment within a pH range of approximately 3 to 14. As the environment becomes more reducing, the concentration of technetium in solution is reduced and the dominant species is $TcO(OH)_{2}^{\circ}$.

If ruthenium migrates, then technetium, which also exists as a negative ion in an oxidizing environment, should also migrate. Seven technetium isotopes are produced in a nuclear detonation.⁹ With the exception of ⁹⁹Tc, none of the technetium isotopes have half-lives that would result in measurable concentrations given the time of the Cambric study. Approximately one-third as many ⁹⁹Tc atoms are produced as stable ruthe-

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nium atoms; however, technetium is not limited by solubility in an oxidizing environment and should be present, albeit in small concentrations. At the present time, the lack of sensitivity in our analytical techniques prevents the study of technetium migration. More sensitive methods are currently being developed by both LLNL and LANL, a coparticipant in the RNM Project. Until the archived samples from the cavity and satellite wells have been analyzed for technetium, we are limited to predictions using geochemical models.

5.6. Summary

Ruthenium migration at the Cambric site was successfully predicted by the model. The solubility of ruthenium in the cavity water was approximately equal to the predicted value. The negative ion predicted as the dominant species has potential for migration. The behavior of ruthenium in laboratory experiments that conflict with the observed behavior of ruthenium can be explained by differences in speciation between laboratory and field conditions. When analytical techniques are improved, there is hope that technetium migration may yet be measured at the Cambric site.

5.7. References

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NMS,JCC/jlc

Appendix A Analytical Methods

This section describes the sample collection, processing, and analytical methods generally used for the RNM well samples. Unique or highly specific methods are described in the appropriate sections.

The salt samples obtained from the evaporation of large-volume water samples (Section 2.1.1) are transferred to a tared container of standard geometry, sealed, weighed, and counted for at least a week on low-background Ge(Li) detector systems. The spectrum is analyzed and results calculated by the GAMANAL computer code.^{1,2} Disk-type filters (e.g., Nuclepore), when used, are counted in a similar fashion. Cartridge or bag filters are cut up and the pieces pressed into the standard containers before sealing and counting. The results are calculated on the basis of the volume of the original water sample. The uncertainties reported are based on counting statistics as calculated by the GAMANAL code; these do not represent the true experimental uncertainties for possibly heterogeneous samples counted in an extended geometry. Based on laboratory trials, the estimated minimum uncertainties are 10–20% for salt samples and 20–30% for filters.

High-activity (chimney or cavity) samples are sometimes counted directly by placing a known aliquot of the water into a standard volume container (Marinelli beaker or Prindle vial).

Tritium analyses are performed by direct liquid scintillation counting of the water sample. The methods employed have been developed and standardized for our environmental tritium laboratory.¹⁻³

During 1983 and 1984, we have systematically developed and standardized our chemical analytical capabilities. We now routinely analyze water samples for the major and minor ionic constituents and, when possible, for standard water quality parameters in the field. Stable cations are analyzed at LLNL by inductively coupled plasma-optical emission spectrometry (ICP-OES) techniques.¹⁻⁴ Anions are analyzed either by ion chromatography or autoanalyzer techniques according to procedures developed and maintained by LLNL's Chemistry Department. In the field, prompt analyses are performed for pH and Eh (data reported vs Normal Hydrogen Electrode) using Orion pH/millivoltmeters and appropriate electrodes, for dissolved oxygen using a Leeds and Northrup DO meter, and for alkalinity, hardness, and sulfides using a Hach field titration kit.

In all cases, care is taken to avoid sample contamination, and all analyses are carried out by calibrated standard methods according to the quality assurance standards adopted by the appropriate LLNL organization.

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Appendix B Soil Moisture Data

Table B1 presents the field data obtained from the soil moisture instrumentation for the Cambric plume study. See Section 4 for a description of the project and experimental site. The key for the table is as follows:

First column: Type of soil moisture instrument. RC = resistance cell, NP = neutron probe, T = tensiometer.

Second column: Date. First two digits are year, last three are day of year.

Third column: Sample# identifies the instrument by number, distance from the berm of the ditch, and depth beneath ground surface. Example: 13,14-5-11 designates resistance cells 13 and 14, both of which are 5 ft from the outer edge of the berm and 11 ft below the surface (see Figs. 4-2 and 4-3). Resistance cells and tensiometers were installed in pairs at each distance and depth; where only one number appears in the first block, it indicates that one of the instruments was not operating.

Fourth column: Reading of instrument (numerical value); where pairs of instruments are operating, the reading reported is the mean of the two readings.

Fifth column: Units of instrument reading—ohms for resistance cells, volume % moisture for the neutron probe, and centibars of soil moisture tension for the tensiometers.

This table has not been edited by comparison with field notes to eliminate readings that may reflect instrument problems or unusual environmental conditions. In particular, many of the neutron probe observations are suspect. "Stars" indicate cases where no reading was obtained (e.g., tensiometers frozen in winter). The authors request that anyone contemplating use or interpretation of these data contact them for a more complete discussion of their significance.

strumentation for		or the Cambi	the Cambric plume study.			D
Туре	Date	Sample#	Reading	Units	Туре	
RC	83192	13,14-5-11	7750	ohms	RC	
RC	83200	13,14-5-11	6900	ohms	RC	
RC	83206	13,14-5-11	6700	ohms	RC	
RC	83213	13,14-5-11	6700	ohms	RC	
RC	83220	13,14-5-11	6250	ohms	RC	
RC	83227	13,14-5-11	6050	ohms	RC	
RC	83241	13,14-5-11	6000	ohms	RC	
RC	83250	13,14-5-11	6000	ohms	RC	
RC	83255	13,14-5-11	6000	ohms	RC	
RC	83262	13,14-5-11	6100	ohms	RC	
RC	83271	13,14-5-11	6100	ohms	RC	
RC	83283	13,14-5-11	6150	ohms	RC	
RC	83297	13,14-5-11	6150	ohms	RC	
RC	83305	13,14-5-11	6400	ohms	RC	
RC	83311	13,14-5-11	5900	ohms	RC	
RC	83318	13,14-5-11	6100	ohms	RC	
RC	83325	13,14-5-11	5850	ohms	RC	
RC	83332	13,14-5-11	6150	ohms	RC	
RC	83339	13,14-5-11	6250	ohms	RC	
RC	83346	13,14-5-11	6250	ohms	RC	
RC	83353	13,14-5-11	6250	ohms	RC	
RC	83361	13,14-5-11	6200	ohms	RC	
RC	84003	13,14-5-11	6350	ohms	RC	
RC	84009	13,14-5-11	6350	ohms	RC	
RC	84016	13,14-5-11	6350	ohms	RC	
RC	84030	13,14-5-11	6700	ohms	RC	
RC	84037	13,14-5-11	6500	ohms	RC	

Table B-1.	Field	data	from	soil	moisture	in-
strumentati	ion for	the (Cambr	ic pl	ume study	

Table B-1. Continued.

Туре	Date	Sample#	Reading	Units
RC	84044	13,14-5-11	7000	ohms
RC	84052	13,14-5-11	6950	ohms
RC	84058	13,14-5-11	7150	ohms
RC	84065	13,14-5-11	7150	ohms
RC	84072	13,14-5-11	6950	ohms
RC	84079	13,14-5-11	7100	ohms
RC	84086	13,14-5-11	7200	ohms
RC	84093	13,14-5-11	7200	ohms
RC	84100	13,14-5-11	7250	ohms
RC	84107	13,14-5-11	7100	ohms
RC	84114	13,14-5-11	7350	ohms
RC	84121	13,14-5-11	7800	ohms
RC	84128	13,14-5-11	7650	ohms
RC	84135	13,14-5-11	7300	ohms
RC	84142	13,14-5-11	7500	ohms
RC	84151	13,14-5-11	7300	ohms
RC	84156	13,14-5-11	7250	ohms
RC	84163	13,14-5-11	7250	ohms
RC	84170	13,14-5-11	7350	ohms
RC	84177	13,14-5-11	7250	ohms
RC	84184	13,14-5-11	7100	ohms
RC	84191	13,14-5-11	7250	ohms
RC	84198	13,14-5-11	7400	ohms
RC	84205	13,14-5-11	7400	ohms
RC	84212	13,14-5-11	7800	ohms
RC	84219	13,14-5-11	7400	ohms
RC	84226	13,14-5-11	6350	ohms

Table B-1. Continued.

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уре	Date	Sample#	Reading	Units	Туре	Date	Sample#	Reading	Uni
RC	83192	22-5-5	7000	ohms	RC	83255	15,16-11-11	3700	ohr
C	83200	22-5-5	6800	ohms	RC	83262	15,16-11-11	3450	ohr
C	83206	22-5-5	6000	ohms	RC	83271	15,16-11-11	3150	ohn
RC	83213	22-5-5	6000	ohms	RC	83283	15,16-11-11	3100	ohr
C	83220	22-5-5	6000	ohms	RC	83297	15,16-11-11	3100	ohr
C	83227	22-5-5	5500	ohms	RC	83305	15,16-11-11	3050	ohr
C	83241	22-5-5	3500	ohms	RC	83311	15,16-11-11	3000 3000	ohi
C	83250	22-5-5	3300	ohms	RC RC	83318 83325	15,16-11-11 15,16-11-11	2900	ohı ohı
C C	83255	22-5-5	3300	ohms	RC	83332	15,16-11-11	2900	ohi
C	83262 83271	22-5-5 22-5-5	3200 3300	ohms ohms	RC	83339	15,16-11-11	2900	oh
IC IC	83283	22-5-5	3300 3400	ohms	RC	83346	15,16-11-11	2900	oh
IC IC	83283	22-5-5	3400	ohms	RC	83353	15,16-11-11	2900	oh
KC	83305	22-5-5	3200	ohms	RC	83361	15,16-11-11	2900	oh
RC	83311	22-5-5	3200	ohms	RC	84003	15,16-11-11	2900	ohi
RC	83318	22-5-5	3200	ohms	RC	84009	15,16-11-11	2900	ohi
IC IC	83325	22-5-5	3200	ohms	RC	84016	15,16-11-11	2900	ohi
iC IC	83332	22-5-5	3400	ohms	RC	84030	15,16-11-11	3150	oh
C	83339	22-5-5	3400	ohms	RC	84037	15,16-11-11	3000	oh
IC .	83346	22-5-5	3500	ohms	RC	84044	15,16-11-11	3050	oh
IC .	83353	22-5-5	3500	ohms	RC	84052	15,16-11-11	3000	oh
C	83361	22-5-5	3600	ohms	RC	84058	15,16-11-11	3050	oh
RC	84003	22-5-5	3600	ohms	RC	84065	15,16-11-11	3100	oh
C	84009	22-5-5	3500	ohms	RC	84072	15,16-11-11	2950	oh
RC	84016	22-5-5	3600	ohms	RC	84079	15,16-11-11	3050	oh
C	84030	22-5-5	3700	ohms	RC	84086	15,16-11-11	3100	oh
C	84037	22-5-5	3500	ohms	RC	84093	15,16-11-11	3000	oh
C	84044	22-5-5	3600	ohms	RC	84100	15,16-11-11	3300	oh
C	84052	22-5-5	3600	ohms	RC	84107	15,16-11-11	3000	oh
C	84058	22-5-5	3600	ohms	RC	84114	15,16-11-11	3000	oh
C	84065	22-5-5	3500	ohms	RC	84121	15,16-11-11	3000	oh
(C	84072	22-5-5	3500	ohms	RC	84128	15,16-11-11	3000	oh
IC .	84079	22-5-5	3400	ohms	RC	84135	15,16-11-11	3000	oh
۲C	84086	22-5-5	3400	ohms	RC	84142	15,16-11-11	2950	oh
RC	84093	22-5-5	3200	ohms	RC	84151	15,16-11-11	2950	oh
RC	84100	22-5-5	3400	ohms	RC	84156	15,16-11-11	2900	oh
RC	84107	22-5-5	3500	ohms	RC	84163	15,16-11-11	2800	oh
۲C	84114	22-5-5	3500	ohms	RC	84170	15,16-11-11	2850	oh
RC .	84121	22-5-5	3500	ohms	RC	84177	15,16-11-11	2800	oh
RC	84128	22-5-5	3500	ohms	RC	84184 84191	15,16-11-11	2600 2750	oh oh
RC	84135	22-5-5	3200	ohms	RC RC	84191 84198	15,16-11-11	2730	oh
RC	84142	22-5-5	3000 3000	ohms ohms	RC	84205	15,16-11-11 15,16-11-11	2800	oh
RC RC	84151 84156	22-5-5 22-5-5	2800	ohms	RC	84203	15,16-11-11	2300	oh
RC	84163	22-5-5	2800	ohms	RC	84219	15,16-11-11	2700	oh
ic ic	84103 84170	22-5-5	2800	ohms	RC	84226	15,16-11-11	2650	oh
IC IC	84170	22-5-5	2800	ohms	RC	83192	18,19-11-5	112500	oh
RC	84184	22-5-5	2700	ohms	RC	83200	18,19-11-5	105000	oh
RC	84191	22-5-5	2300	ohms	RC	83206	18,19-11-5	95000	oh
IC IC	84198	22-5-5	2700	ohms	RC	83213	18,19-11-5	90000	oh
RC	84205	22-5-5	2700	ohms	RC	83220	18,19-11-5	82500	oh
RC	84212	22-5-5	2500	ohms	RC	83227	18,19-11-5	110000	oh
iC	84219	22-5-5	2500	ohms	RC	83241	18,19-11-5	74000	oh
RC	84226	22-5-5	2400	ohms	RC	83250	18,19-11-5	72500	oh
RC	83192	15,16-11-11	20400	ohms	RC	83255	18,19-11-5	72500	oh
RC	83200	15,16-11-11	11650	ohms	RC	83262	18,19-11-5	67000	oh
RC	83206	15,16-11-11	10400	ohms	RC	83271	18,19-11-5	67500	oh
RC	83213	15,16-11-11	6550	ohms	RC	83283	18,19-11-5	67500	oh
RC	83220	15,16-11-11	6250	ohms	RC	83297	18,19-11-5	65000	oh
IC	83227	15,16-11-11	4800	ohms	RC	83305	18,19-11-5	62000	oh
iC	83241	15,16-11-11	3950	ohms	RC	83311	18,19-11-5	64500	oh
RC	83250	15,16-11-11	3750	ohms	RC	83318	18,19-11-5	62000	oh

Table B-1. Continued.

Table B-1. Continued.

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Units ohms ohms

Table B-1. Continued.					Table B-1. Continued.				
Туре	Date	Sample#	Reading	Units	Туре	Date	Sample#	Reading	
RC	83325	18,19-11-5	62500	ohms	RC	84016	9,12-11-7	4400	
С	83332	18,19-11-5	62500	ohms	RC	84030	9,12-11-7	4750	
	83339	18,19-11-5	62500	ohms	RC	84037	9,12-11-7	5000	
	83346	18,19-11-5	62000	ohms	RC	84044	9,12-11-7	4800	
	83353	18,19-11-5	61500	ohms	RC	84052	9,12-11-7	4850	
	83361	18,19-11-5	60500	ohms	RC	84058	9,12-11-7	5100	
	84003	18,19-11-5	62500	ohms	RC	84065	9,12-11-7	4850	
	84009	18,19-11-5	61500	ohms	RC	84072	9,12-11-7	4850	
	84016	18,19-11-5	59750	ohms	RC	84079	9,12-11-7	4900	
	84030	18,19-11-5	63500	ohms	RC	84086	9,12-11-7	4850	
2	84037	18,19-11-5	62500	ohms	RC	84093	9,12-11-7	4900	
	84044	18,19-11-5	62500	ohms	RC	84100	9,12-11-7	4950	
2	84052	18,19-11-5	63000	ohms	RC	84107	9,12-11-7	5050	
	84058	18,19-11-5	62500	ohms	RC	84114	9,12-11-7	4900	
	84065	18,19-11-5	62500	ohms	RC	84121	9,12-11-7	5100	
	84072	18,19-11-5	61500	ohms	RC	84128	9,12-11-7	5150	
	84079	18,19-11-5	61500	ohms	RC	84135	9,12-11-7	5150	
	84086	18,19-11-5	60000	ohms	RC	84142	9,12-11-7	4850	
	84093	18,19-11-5	59500	ohms	RC	84151	9,12-11-7	4950	
	84100	18,19-11-5	59500	ohms	RC	84156	9,12-11-7	4800	
	84107	18,19-11-5	58500	ohms	RC	84163	9,12-11-7	4750	
2	84114	18,19-11-5	55500	ohms	RC	84170	9,12-11-7	4900	
-	84121	18,19-11-5	57500	ohms	RC	84177	9,12-11-7	4750	
2	84128	18,19-11-5	56500	ohms	RC	84184	9,12-11-7	4750	
	84135	18,19-11-5	52500	ohms	RC	84191	9,12-11-7	4750	
	84142	18,19-11-5	52000	ohms	RC	84198	9,12-11-7	4750	
	84151	18,19-11-5	52500	ohms	RC	84205	9,12-11-7	4750	
	84156	18,19-11-5	51500	ohms	RC	84212	9,12-11-7	4750	
	84163	18,19-11-5	49500	ohms	RC	84219	9,12-11-7	4850	
	84170	18,19-11-5	51500	ohms	RC	84226	9,12-11-7	4800	
	84177	18,19-11-5	53000	ohms	RC	83192	24,25-11-3	1150000	
	84184	18,19-11-5	53000	ohms	RC	83200	24,25-11-3	1200000	
	84191	18,19-11-5	48000	ohms	RC	83206	24,25-11-3	1125000	
	84198	18,19-11-5	50000	ohms	RC	83213	24,25-11-3	1125000	
	84205	18,19-11-5	45000	ohms	RC	83220	24,25-11-3	825000	
	84212	18,19-11-5	43000	ohms	RC	83227	24,25-11-3	80000	
-	84219	18,19-11-5	42000	ohms	RC	83241	24,25-11-3	800000	
2	84226	18,19-11-5	38500	ohms	RC	83250	24,25-11-3	550000	
C	83192	9,12-11-7	4500	ohms	RC	83255	24,25-11-3	450000	
2	83200	9,12-11-7	4100	ohms	RC	83262	24,25-11-3	240000	
2	83206	9,12-11-7	3700	ohms	RC	83271	24,25-11-3	112500	
2	83213	9,12-11-7	3550	ohms	RC	83283	24,25-11-3	107000	
	83220	9,12-11-7 9,12-11-7	3500 3400	ohms ohms	RC	83297 83305	24,25-11-3 24,25-11-3	115000	
	83227 83241	•	3400	ohms	RC RC	83305 83311	24,25-11-3 24,25-11-3	107500	
C C	83241	9,12-11-7						110000	
	83250	9,12-11-7 9,12-11-7	3200 3200	ohms	RC	83318 83325	24,25-11-3	122500	
C	83255	9,12-11-7	3200 3150	ohms	RC	83325 83332	24,25-11-3 24,25-11-3	125000	
C C	83262 83271	9,12-11-7 9,12-11-7	3150 3150	ohms ohms	RC RC	83332 83339	24,25-11-3 24,25-11-3	140000 145000	
	83283	9,12-11-7 9,12-11-7	3150	ohms	RC	83346	24,25-11-3		
-								160000	
	83297 83305	9,12-11-7 9,12-11-7	3450 3450	ohms ohms	RC RC	83353 83361	24,25-11-3 24,25-11-3	145000 155000	
-		9,12-11-7 9,12-11-7	3450		RC	84003			
	83311 83318		3450	ohms ohms		84003 84009	24,25-11-3 24,25-11-3	155000 145000	
2	83318	9,12-11-7 9,12-11-7		ohms	RC RC		24,25-11-3		
C C	83325	9,12-11-7 9,12-11-7	3600 3700	ohms	RC	84016 84030	24,25-11-3 24,25-11-3	147500	
C C	83332	9,12-11-7		ohms			24,25-11-3	145000	
	83339	9,12-11-7 9,12-11-7	3800	ohms	RC	84037 84044	24,25-11-3	140000	
2	83346	9,12-11-7 9,12-11-7	3950 3950	ohms	RC	84044 84052	24,25-11-3	130000	
	83353	9,12-11-7	3950	ohms	RC	84052	24,25-11-3	135000	
	83361	9,12-11-7	4200	ohms	RC	84058	24,25-11-3	130000	
	04003	0 10 11 7	4400	ahma	D D C	840CE	34 35 11 3	10-000	
.C .C .C	84003 84009	9,12-11-7 9,12-11-7	4400 4350	ohms ohms	RC RC	84065 84072	24,25-11-3 24,25-11-3	125000 125000	

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Table B-1. Continued.

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Table B-1. Continued.

Units ohms ohms

Гуре	Date	Sample#	Reading	Units	Type	Date	Sample#	Reading
RC	84079	24,25-11-3	112500	ohms	RC	84135	17,20-21-4	875000
RC	84086	24,25-11-3	110000	ohms	RC	84142	17,20-21-4	875000
С	84093	24,25-11-3	109000	ohms	RC	84151	17,20-21-4	900000
2	84100	24,25-11-3	104000	ohms	RC	84156	17,20-21-4	875000
	84107	24,25-11-3	94000	ohms	RC	84163	17,20-21-4	900000
	84114	24,25-11-3	95000	ohms	RC	84170	17,20-21-4	1000000
	84121	24,25-11-3	95000	ohms	RC	84177	17,20-21-4	900000
	84128	24,25-11-3	94000	ohms	RC	84184	17,20-21-4	1250000
	84135	24,25-11-3	86500	ohms	RC	84191	17,20-21-4	1150000
2	84142	24,25-11-3	86000	ohms	RC	84198	17,20-21-4	1150000
С	84151	24,25-11-3	83500	ohms	RC	84205	17,20-21-4	1150000
С	84156	24,25-11-3	89000	ohms	RC	84212	17,20-21-4	1150000
С	84163	24,25-11-3	94000	ohms	RC	84219	17,20-21-4	1000000
С	84170	24,25-11-3	102500	ohms	RC	84226	17,20-21-4	1150000
С	84177	24,25-11-3	110000	ohms	RC	83192	30,31-21-10	160000
C	84184	24,25-11-3	115000	ohms	RC	83200	30,31-21-10	155000
С	84191	24,25-11-3	120000	ohms	RC	83206	30,31-21-10	140000
С	84198	24,25-11-3	117500	ohms	RC	83213	30,31-21-10	135000
С	84205	24,25-11-3	117500	ohms	RC	83220	30,31-21-10	135000
С	84212	24,25-11-3	110000	ohms	RC	83227	30,31-21-10	120000
C	84219	24,25-11-3	105000	ohms	RC	83241	30,31-21-10	115000
C	84226	24,25-11-3	96000	ohms	RC	83250	30,31-21-10	110000
RC	83192	17,20-21-4	1400000	ohms	RC	83255	30,31-21-10	110000
c	83200	17,20-21-4	1600000	ohms	RC	83262	30,31-21-10	102500
ĸĊ	83206	17,20-21-4	1750000	ohms	RC	83271	30,31-21-10	100000
C	83213	17,20-21-4	1750000	ohms	RC	83283	30,31-21-10	97500
C	83220	17,20-21-4	750000	ohms	RC	83297	30,31-21-10	92500
Ĉ	83227	17,20-21-4	750000	ohms	RC	83305	30,31-21-10	90000
C	83241	17,20-21-4	1500000	ohms	RC	83311	30,31-21-10	90000
2	83250	17,20-21-4	1750000	ohms	RC	83318	30,31-21-10	90000
	83255	17,20-21-4	1500000	ohms	RC	83325	30,31-21-10	90000
	83262	17,20-21-4	1250000	ohms	RC	83332	30,31-21-10	87500
	83271	17,20-21-4	1250000	ohms	RC	83339	30,31-21-10	87500
	83283	17,20-21-4	1500000	ohms	RC	83346	30,31-21-10	90000
	83297	17,20-21-4	1500000	ohms	RC	83353	30,31-21-10	90000
-	83305	17,20-21-4	1250000	ohms	RC	83361	30,31-21-10	89500
2	83311	17,20-21-4	1250000	ohms	RC	84003	30,31-21-10	89500
C	83318	17,20-21-4	1250000	ohms	RC	84009	30,31-21-10	90000
c	83325	17,20-21-4	900000	ohms	RC	84016	30,31-21-10	90000
c	83332	17,20-21-4	875000	ohms	RC	84030	30,31-21-10	92500
C	83339	17,20-21-4	900000	ohms	RC	84037	30,31-21-10	90000
C	83346	17,20-21-4	900000	ohms	RC	84044	30,31-21-10	90000
C	83353	17,20-21-4	875000	ohms	RC	84052	30,31-21-10	95000
C	83361	17,20-21-4	1000000	ohms	RC	84058	30,31-21-10	95000
c c	84003	17,20-21-4	875000	ohms	RC	84065	30,31-21-10	95000
c	84003 84009		1125000	ohms	RC	84072	30,31-21-10	87500
		17,20-21-4 17,20-21-4		ohms	RC	84072	30,31-21-10	90000
C	84016		875000			84079 84086	30,31-21-10	90000
C	84030	17,20-21-4	1250000	ohms	RC		,	90000
C	84037	17,20-21-4	1000000	ohms	RC RC	84093	30,31-21-10	
C	84044	17,20-21-4	1200000	ohms	RC	84100	30,31-21-10	92500
C	84052	17,20-21-4	1050000	ohms	RC	84107	30,31-21-10	90000
C	84058	17,20-21-4	1200000	ohms	RC	84114	30,31-21-10	90000
C	84065	17,20-21-4	875000	ohms	RC	84121	30,31-21-10	89000
C	84072	17,20-21-4	875000	ohms	RC	84128	30,31-21-10	86500
IC .	84079	17,20-21-4	875000	ohms	RC	84135	30,31-21-10	85000
C	84086	17,20-21-4	875000	ohms	RC	84142	30,31-21-10	84000
C	84093	17,20-21-4	875000	ohms	RC	84151	30,31-21-10	84000
С	84100	17,20-21-4	875000	ohms	RC	84156	30,31-21-10	83500
С	84107	17,20-21-4	875000	ohms	RC	84163	30,31-21-10	80000
2	84114	17,20-21-4	800000	ohms	RC	84170	30,31-21-10	82500
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2	84121	17,20-21-4	875000	ohms	RC	84177	30,31-21-10	79000

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Table B-1. Continued.

Table B-1. Continued.

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	able B-1. Continued.					Table B-1. Continued.					
Туре	Date	Sample#	Reading	Units	Type	Date	Sample#	Reading	Unit		
RC	84191	30,31-21-10	74000	ohms	RC	83206	7,8-11-10	7500	ohm		
RC	84198	30,31-21-10	75000	ohms	RC	83213	7,8-11-10	7000	ohm		
RC	84205	30,31-21-10	74000	ohms	RC	83220	7,8-11-10	6050	ohm		
RC	84212	30,31-21-10	70000	ohms	RC	83227	7,8-11-10	5900	ohm		
RC	84219	30,31-21-10	68500	ohms	RC	83241	7,8-11-10	5250	ohm		
RC	84226	30,31-21-10	66500	ohms	RC	83250	7,8-11-10	5100	ohm		
RC	83192	1,2-11-15	660	ohms	RC	83255	7,8-11-10	5000	ohm		
RC	83200	1,2-11-15	1010	ohms	RC	83262	7,8-11-10	6000	ohm		
RC	83206	1,2-11-15	1250	ohms	RC	83271	7,8-11-10	5000	ohms		
RC	83213	1,2-11-15	1450	ohms	RC	83283	7,8-11-10	4650	ohm		
RC	83220	1,2-11-15	1550	ohms	RC	83297	7,8-11-10	4650	ohm		
RC	83227	1,2-11-15	1700	ohms	RC	83305	7,8-11-10	4650	ohm		
RC	83241	1,2-11-15	1725	ohms	RC	83311	7,8-11-10	4650	ohm		
RC	83250	1,2-11-15	1740	ohms	RC	83318	7,8-11-10	4650	ohm		
RC	83255	1,2-11-15	1790	ohms	RC	83325	7,8-11-10	4450	ohm		
RC	83262	1,2-11-15	1800	ohms	RC	83332	7,8-11-10	4300	ohm		
RC	83271	1,2-11-15	1800	ohms	RC	83339	7,8-11-10	4300	ohm		
RC	83283	1,2-11-15	1900	ohms	RC	83346	7,8-11-10	4400	ohm		
RC	83297	1,2-11-15	1900	ohms	RC	83353	7,8-11-10	4300	ohm		
RC	83305	1,2-11-15	1950	ohms	RC	83361	7,8-11-10	4450	ohm		
RC	83311	1,2-11-15	1850	ohms	RC	84003	7,8-11-10	4450	ohm		
RC	83318	1,2-11-15	1900	ohms	RC	84009	7,8-11-10	4500	ohm		
RC	83325	1,2-11-15	1400	ohms	RC	84016	7,8-11-10	4500	ohm		
RC	83332	1,2-11-15	1850	ohms	RC	84030	7,8-11-10	4550	ohm		
RC	83339	1,2-11-15	1900	ohms	RC	84037	7,8-11-10	4550	ohm		
RC	83346	1,2-11-15	1950	ohms	RC	84044	7,8-11-10	4925	ohm		
RC	83353	1,2-11-15	1950	ohms	RC	84052	7,8-11-10	4800	ohm		
RC	83361	1,2-11-15	1875	ohms	RC	84058	7,8-11-10	4850	ohm		
RC	84003	1,2-11-15	1875	ohms	RC	84065	7,8-11-10	4900	ohm		
RC	84009	1,2-11-15	1925	ohms	RC	84072	7,8-11-10	4850	ohm		
RC	84016	1,2-11-15	1925	ohms	RC	84072	7,8-11-10	4900	ohm		
RC	84030	1,2-11-15	2100	ohms	RC	84086	7,8-11-10	4850	ohm		
RC	84030	1,2-11-15	2050	ohms	RC	84093	7,8-11-10	4700	ohm		
					RC	84093 84100		5150	ohm		
RC	84044	1,2-11-15	2013	ohms		84100 84107	7,8-11-10	4900	ohm		
RC	84052	1,2-11-15	2200	ohms	RC		7,8-11-10				
RC	84058	1,2-11-15	2200	ohms	RC	84114	7,8-11-10	5100	ohm		
RC	84065	1,2-11-15	2400	ohms	RC	84121	7,8-11-10	5300	ohm		
RC	84072	1,2-11-15	2350	ohms	RC	84128	7,8-11-10	5000	ohm		
RC	84079	1,2-11-15	2300	ohms	RC	84135	7,8-11-10	4800	ohm		
RC	84086	1,2-11-15	2300	ohms	RC	84142	7,8-11-10	4900	ohm		
RC	84093	1,2-11-15	2300	ohms	RC	84151	7,8-11-10	4800	ohm		
RC	84100	1,2-11-15	2350	ohms	RC	84156	7,8-11-10	4650	ohm		
RC	84107	1,2-11-15	2450	ohms	RC	84163	7,8-11-10	4650	ohm		
RC	84114	1,2-11-15	2450	ohms	RC	84170	7,8-11-10	4550	ohm		
RC	84121	1,2-11-15	2450	ohms	RC	84177	7,8-11-10	4550	ohm		
RC	84128	1,2-11-15	2350	ohms	RC	84184	7,8-11-10	4500	ohm		
RC	84135	1,2-11-15	2450	ohms	RC	84191	7,8-11-10	4750	ohm		
RC	84142	1,2-11-15	2450	ohms	RC	84198	7,8-11-10	4400	ohm		
RC	84151	1,2-11-15	2450	ohms	RC	84205	7,8-11-10	4400	ohm		
RC	84156	1,2-11-15	2450	ohms	RC	84212	7,8-11-10	4250	ohn		
RC	84163	1,2-11-15	2450	ohms	RC	84219	7,8-11-10	4250	ohn		
RC	84170	1,2-11-15	2350	ohms	RC	84226	7,8-11-10	4050	ohm		
RC	84177	1,2-11-15	2450	ohms	RC	83192	10,11-11-5	230000	ohm		
RC	84184	1,2-11-15	2400	ohms	RC	83200	10,11-11-5	235000	ohm		
RC	84191	1,2-11-15	2450	ohms	RC	83206	10,11-11-5	250000	ohm		
RC	84198	1,2-11-15	2550	ohms	RC	83213	10,11-11-5	235000	ohm		
RC	84205	1,2-11-15	3000	ohms	RC	83220	10,11-11-5	18500	ohm		
RC	84212	1,2-11-15	2450	ohms	RC	83227	10,11-11-5	17000	ohm		
RC	84219	1,2-11-15	2450	ohms	RC	83241	10,11-11-5	145000	ohm		
RC	84226	1,2-11-15	2450	ohms	RC	83250	10,11-11-5	140000	ohm		
	83192	7,8-11-10	9000	ohms	RC	83255	10,11-11-5	130000	ohm		
RC											

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Table B-1. Continued.

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Units %VM %VM%VM %VM %VM%VM %VM % VM%VM %VM %VM % VM%VM %VM %VM %VM %VM %VM %VM %VM %VM %VM %VM

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Гуре	Date	Sample#	Reading	Units	Туре	Date	Sample#	Reading
RC	83271	10,11-11-5	115000	ohms	NP	83339	11-5	13.1
۲C	83283	10,11-11-5	110000	ohms	NP	83346	11-5	13.2
RC	83297	10,11-11-5	105000	ohms	NP	83353	11-5	12.7
RC	83305	10,11-11-5	100000	ohms	NP	83361	11-5	13.0
RC	83311	10,11-11-5	100000	ohms	NP	84003	11-5	12.5
RC	83318	10,11-11-5	100000	ohms	NP	84009	11-5	13.2
RC	83325	10,11-11-5	100000	ohms	NP	84016	11-5	13.5
RC	83332	10,11-11-5	100000	ohms	NP	84030	11-5	****
RC	83339	10,11-11-5	100000	ohms	NP	84037	11-5	****
RC	83346	10,11-11-5	100000	ohms	NP	84044	11-5	****
RC	83353	10,11-11-5	97500	ohms	NP	84052	11-5	****
RC	83361	10,11-11-5	100000	ohms	NP	84058	11-5	****
RC	84003	10,11-11-5	99500	ohms		84065	11-5	****
RC	84009	10,11-11-5	105000	ohms	NP	84072	11-5	****
RC	84016	10,11-11-5	97500	ohms	NP	84079 84086	11-5	****
RC	84030	10,11-11-5	104500	ohms		84086	11-5	****
RC	84037	10,11-11-5	101500	ohms	NP NP	84093	11-5 11-5	
RC	84044	10,11-11-5	100000	ohms		84100		12.8
RC	84052	10,11-11-5	100000	ohms	NP	84107	11-5	12.1
RC	84058	10,11-11-5	100000	ohms	NP NP	84114	11-5 11-5	12.3 13.3
RC	84065	10,11-11-5	100000	ohms	NP	84121 84128	11-5	13.3
RC RC	84072 84079	10,11-11-5 10,11-11-5	89000 89000	ohms ohms	NP	84128 84135	11-5	12.8
RC	84079	10,11-11-5	88500	ohms	NP	84133	11-5	13.2
RC	84093	10,11-11-5	83500	ohms	NP	84151	11-5	****
RC	84100	10,11-11-5	83000	ohms	NP	84156	11-5	****
RC	84100 84107	10,11-11-5	78000	ohms	NP	84163	11-5	****
RC	84114	10,11-11-5	75000	ohms	NP	84170	11-5	****
RC	84121	10,11-11-5	73500	ohms	NP	84177	11-5	****
RC	84128	10,11-11-5	68500	ohms	NP	84184	11-5	****
RC	84135	10,11-11-5	66000	ohms	NP	84191	11-5	****
RC	84142	10,11-11-5	62500	ohms	NP	84198	11-5	13.6
RC	84151	10,11-11-5	58000	ohms	NP	84205	11-5	13.8
RC	84156	10,11-11-5	56500	ohms	NP	84212	11-5	****
RC	84163	10,11-11-5	54500	ohms	NP	84219	11-5	12.9
RC	84170	10,11-11-5	51000	ohms	NP	84226	11-5	13.0
RC	84177	10,11-11-5	47500	ohms	NP	83192	11-7.5	15.2
RC	84184	10,11-11-5	45000	ohms	NP	83200	11-7.5	15.5
RC	84191	10,11-11-5	42500	ohms	NP	83206	11-7.5	15.1
RC	84198	10,11-11-5	37500	ohms	NP	83213	11-7.5	14.7
RC	84205	10,11-11-5	37500	ohms	NP	83220	11-7.5	15.5
RC	84212	10,11-11-5	36000	ohms	NP	83227	11-7.5	14.7
RC	84219	10,11-11-5	36000	ohms	NP	83241	11-7.5	14.8
RC	84226	10,11-11-5	33000	ohms	NP	83250	11-7.5	14.9
NP	83192	11-5	12.00	%VM	NP	83255	11-7.5	15.3
NP	83200	11-5	12.00	%VM	NP	83262	11-7.5	13.0
NP	83206	11-5	12.7	%VM	NP	83271	11-7.5	14.9
NP	83213	11-5	12.3	%VM	NP	83283	11-7.5	14.3
NP	83220	11-5	13.0	%VM	NP	83297	11-7.5	14.4
NP	83227	11-5	12.5	%VM	NP	83305	11-7.5	14.6
NP	83241	11-5	13.3	%VM	NP	83311	11-7.5	14.3
NP	83250	11-5	13.0	%VM	NP	83318	11-7.5	14.4
NP	83255	11-5	13.1	%VM	NP	83325	11-7.5	14.5
NP	83262	11-5	15.2	%VM	NP	83332	11-7.5	14.6
NP	83271	11-5	13.0	%VM	NP	83339	11-7.5	14.3
NP	83283	11-5	13.7	%VM	NP	83346	11-7.5	15.0
NP	83297	11-5	13.0	%VM	NP	83353	11-7.5	14.2
NP	83305	11-5	14.0	%VM	NP	83361	11-7.5	14.2
NP	83311	11-5	12.9	%VM	NP	84003	11-7.5	14.6
NP	83318	11-5	13.6	%VM	NP	84009	11-7.5	14.8
NP	83325	11-5	13.1	%VM	NP	84016	11-7.5	14.8 ****
NP	83332	11-5	13.1	%VM	NP	84030	11-7.5	***

Table B-1. Continued.

Table B-1. Continued.

Units

%VM %VM%VM %VM %VM%VM %VM %VM %VM %VM%VM%VM %VM %VM %VM %VM %VM %VM

Table B-1. Continued.					Table B-1. Continued.				
Туре	Date	Sample#	Reading	Units	Type	Date	Sample#	Readin	
NP	84037	11-7.5	****	%VM	NP	84093	11-10	****	
NP	84044	11-7.5	****	%VM	NP	84100	11-10	14.5	
NP	84052	11-7.5	* * * *	%VM	NP	84107	11-10	13.7	
NP	84058	11-7.5	****	%VM	NP	84114	11-10	13.9	
NP	84065	11-7.5	****	%VM	NP	84121	11-10	13.4	
NP	84072	11-7.5	****	%VM	NP	84128	11-10	14.0	
NP	84079	11-7.5	****	%VM	NP	84135	11-10	13.6	
NP	84086	11-7.5	****	%VM	NP	84142	11-10	13.3	
NP	84093	11-7.5	****	%VM	NP	84151	11-10	****	
NP	84100	11-7.5	15.5	%VM	NP	84156	11-10	****	
NP	84107	11-7.5	14.8	%VM	NP	84163	11-10	****	
NP	84114	11-7.5	14.6	%VM	NP	84170	11-10	****	
NP	84121	11-7.5	15.3	%VM	NP	84177	11-10	****	
NP	84128	11-7.5	15.7	%VM	NP	84184	11-10	****	
NP	84135	11-7.5	14.4	%VM	NP	84191	11-10	****	
NP	84142	11-7.5	15.3	%VM	NP	84198	11-10	13.2	
NP	84151	11-7.5	****	%VM	NP	84205	11-10	14.4	
NP	84156	11-7.5	****	%VM	NP	84212	11-10	****	
NP	84163	11-7.5	****	%VM	NP	84219	11-10	13.1	
NP	84170	11-7.5	****	%VM	NP	84226	11-10	11.1	
NP	84177	11-7.5	****	%VM	NP	83192	11-12.5	14.6	
NP	84184	11-7.5	****	%VM	NP	83200	11-12.5	14.7	
NP	84191	11-7.5	****	%VM	NP	83206	11-12.5	15.5	
NP	84198	11-7.5	15.1	%VM	NP	83213	11-12.5	14.5	
NP	84205	11-7.5	14.7	%VM	NP	83220	11-12.5	15.0	
NP	84212	11-7.5	****	%VM	NP	83227	11-12.5	14.8	
NP	84219	11-7.5	16.3	%VM	NP	83241	11-12.5	14.5	
NP	84226	11-7.5	13.3	%VM	NP	83250	11-12.5	15.1	
NP	83192	11-10	14.8	%VM	NP	83255	11-12.5	14.4	
NP	83200	11-10	15.0	%VM	NP	83262	11-12.5	14.0	
NP	83206	11-10	14.8	%VM	NP	83271	11-12.5	14.8	
NP	83213	11-10	14.6	%VM	NP	83283	11-12.5	14.2	
NP	83220	11-10	15.3	%VM	NP	83297	11-12.5	15.4	
NP	83227	11-10	14.5	%VM %VM	NP	83305	11-12.5	14.3	
NP	83241	11-10	14.7		NP	83311	11-12.5	14.2	
NP	83250 83255	11-10	14.7	%VM	NP	83318	11-12.5	15.6	
NP NP	83262	11-10 11-10	13.3 15.1	%VM %VM	NP NP	83325 83332	11-12.5 11-12.5	14.5 14.3	
NP	83271	11-10	13.7	% V M	NP	83339	11-12.5	14.5	
NP	83283	11-10	14.6	% VM	NP	83346	11-12.5	14.0	
NP	83297	11-10	14.0	%VM	NP	83353	11-12.5	13.0	
NP	83305	11-10	13.3	%VM	NP	83361	11-12.5	14.6	
NP	83311	11-10	13.7	%VM	NP	84003	11-12.5	14.0	
NP	83318	11-10	14.8	%VM	NP	84009	11-12.5	14.5	
NP	83325	11-10	13.2	%VM	NP	84016	11-12.5	15.0	
NP	83332	11-10	14.4	%VM	NP	84030	11-12.5	****	
NP	83339	11-10	15.3	%VM	NP	84037	11-12.5	****	
NP	83346	11-10	14.8	%VM	NP	84044	11-12.5	****	
NP	83353	11-10	13.6	% VM	NP	84052	11-12.5	****	
NP	83361	11-10	14.2	%VM	NP	84058	11-12.5	****	
NP	84003	11-10	14.0	%VM	NP	84065	11-12.5	****	
NP	84009	11-10	14.0	%VM ·	NP	84072	11-12.5	****	
NP	84016	11-10	14.3	%VM	NP	84079	11-12.5	****	
NP	84030	11-10	****	%VM	NP	84086	11-12.5	****	
NP	84037	11-10	****	%VM	NP	84093	11-12.5	****	
NP	84037	11-10	****	% VM	NP	84093 84100	11-12.5	14.9	
NP	84052	11-10	***	%VM	NP	84100	11-12.5	14.9	
NP	84052 84058	11-10	***	%VM	NP	84114	11-12.5	13.1	
NP	84058 84065	11-10	****	%VM	NP	84121	11-12.5	13.9	
NP	84083 84072	11-10	****	%VM	NP	84121 84128	11-12.5	14.8	
NP NP	84072 84079	11-10	****	% V M % V M	NP	84135	11-12.5	15.1	
	010/9	11 10		/U V IVI	1 1 1 1	01100	11-14.5	13.1	

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Table B-1. Continued.

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Table D-1. Commune	le B-1. Cor	ıtinued.
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Units

%VM %VM %VM %VM cbar cbar

Sample# 11-12.5 11-12.5 11-12.5 11-12.5 11-12.5 11-12.5 11-12.5 11-12.5 11-12.5 11-12.5 11-12.5 11-12.5 11-12.5 11-12.5 11-15	Reading **** **** **** **** 14.7 14.6 **** 14.1 12.5 14.0 14.2 13.5 13.0	Units %VM %VM %VM %VM %VM %VM %VM %VM %VM %VM	Type NP NP NP T T T T T T T T	Date 84205 84212 84219 84226 83192 83200 83206 83213 83220 83227	Sample# 11-15 11-15 11-15 11-15 1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3	*** 12 1(2 8 9 8
11-12.5 $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-15$	**** **** **** 14.7 14.6 **** 14.1 12.5 14.0 14.2 13.5	%VM %VM %VM %VM %VM %VM %VM %VM %VM %VM	NP NP T T T T T T	84212 84219 84226 83192 83200 83206 83213 83220 83227	11-15 11-15 11-15 1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3	12 10 7 8 9 8
11-12.5 $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-15$	**** **** **** 14.7 14.6 **** 14.1 12.5 14.0 14.2 13.5	%VM %VM %VM %VM %VM %VM %VM %VM %VM	NP NP T T T T T T	84219 84226 83192 83200 83206 83213 83220 83227	11-15 11-15 1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3	7 8 9 8
11-12.5 $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-15$	**** **** 14.7 14.6 **** 14.1 12.5 14.0 14.2 13.5	%VM %VM %VM %VM %VM %VM %VM %VM	NP T T T T T T	84226 83192 83200 83206 83213 83220 83227	11-15 1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3	10 7 8 9 8
11-12.5 $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-15$	**** **** 14.7 14.6 **** 14.1 12.5 14.0 14.2 13.5	%VM %VM %VM %VM %VM %VM %VM	T T T T T T	83192 83200 83206 83213 83220 83227	1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3	10 7 8 9 8
11-12.5 $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-15$	**** 14.7 14.6 **** 14.1 12.5 14.0 14.2 13.5	%VM %VM %VM %VM %VM %VM %VM	T T T T T	83200 83206 83213 83220 83227	1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3	8 9 8
11-12.5 $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-15$	**** 14.7 14.6 **** 14.1 12.5 14.0 14.2 13.5	%VM %VM %VM %VM %VM %VM	T T T T T	83200 83206 83213 83220 83227	1,6-0-3 1,6-0-3 1,6-0-3 1,6-0-3	8 9 8
11-12.5 $11-12.5$ $11-12.5$ $11-12.5$ $11-12.5$ $11-15$	14.7 14.6 **** 14.1 12.5 14.0 14.2 13.5	%VM %VM %VM %VM %VM	T T T T	83206 83213 83220 83227	1,6-0-3 1,6-0-3 1,6-0-3	9 8
11-12.5 $11-12.5$ $11-12.5$ $11-12.5$ $11-15$	14.6 **** 14.1 12.5 14.0 14.2 13.5	%VM %VM %VM %VM	T T T	83213 83220 83227	1,6-0-3 1,6-0-3	8
11-12.5 $11-12.5$ $11-12.5$ $11-12.5$ $11-15$	**** 14.1 12.5 14.0 14.2 13.5	%VM %VM %VM %VM	T T	83220 83227	1,6-0-3	
11-12.5 11-12.5 11-15 11-15 11-15 11-15 11-15 11-15 11-15 11-15	14.1 12.5 14.0 14.2 13.5	%VM %VM %VM %VM	Т	83227		
11-12.5 11-15 11-15 11-15 11-15 11-15 11-15 11-15 11-15	12.5 14.0 14.2 13.5	%VM %VM				8
11-12.5 11-15 11-15 11-15 11-15 11-15 11-15 11-15 11-15	14.0 14.2 13.5	%VM		00041	1,6-0-3	8
11-15 11-15 11-15 11-15 11-15 11-15 11-15	14.0 14.2 13.5	%VM		83241	1,6-0-3	7
11-15 11-15 11-15 11-15 11-15 11-15 11-15	14.2 13.5	%VM	T	83250	1,6-0-3	8
11-15 11-15 11-15 11-15 11-15	13.5		T	83255	1,6-0-3	8
11-15 11-15 11-15 11-15		%VM	T	83262	1,6-0-3	8
11-15 11-15 11-15		%VM	Т	83271	1,6-0-3	7
11-15 11-15	13.8	%VM	Т	83283	1,6-0-3	7
11-15	13.3	%VM	Т	83297	1,6-0-3	7
	14.4	%VM	Т	83305	1,6-0-3	8
11 10	14.7	%VM	Т	83311	1,6-0-3	7
11-15	14.1	%VM	Т	83318	1,6-0-3	7
11-15	13.9	%VM	Т	83325	1,6-0-3	8
11-15	13.9	%VM	Т	83332	1,6-0-3	0
11-15	14.5	%VM	Т	83339	1,6-0-3	*
11-15	14.4	%VM	Т	83346	1,6-0-3	*
11-15	14.5	%VM	Т	83353	1,6-0-3	*
11-15	14.5	% VM	Т	83361	1,6-0-3	*
11-15	13.9	%VM	Т	84003	1,6-0-3	*
		% V M	Т	84009	1,6-0-3	*
11-15	13.8	%VM %VM	Т	84016	1,6-0-3	*
11-15	13.7		Т	84030	1,6-0-3	*
11-15	13.3	%VM	Т	84037	1,6-0-3	*
11-15	12.9	%VM	Т	84044	1,6-0-3	*
11-15	13.7	%VM	Т	84052	1,6-0-3	*
11-15	14.1	%VM	Т	84058	1,6-0-3	*
11-15	13.0	%VM	Т	84065	1,6-0-3	*
11-15	13.9	%VM	Т	84072	1,6-0-3	*
11-15	13.7	%VM	Т	84079	1.6-0-3	0
11-15	****	%VM	Т	84086	1,6-0-3	0
11-15	****	%VM	T	84093	1,6-0-3	0
11-15	****	%VM	Т	84100	1,6-0-3	0
11-15		%VM	Т	84107	1,6-0-3	0
11-15	****	%VM	Т	84114	1,6-0-3	0
11-15	***	%VM	Т	84121	1,6-0-3	0
11-15	****	%VM	Т	84128	1,6-0-3	0
11-15	***	%VM	Ť	84135	1,6-0-3	0
11-15	****	%VM	T	84142	1,6-0-3	0
11-15	****	%VM	T	84151	1,6-0-3	0
11-15	****	%VM	T	84156	1,6-0-3	0
11-15	13.6	%VM	T	84163	1,6-0-3	0
11-15	13.7	%VM	T	84170	1,6-0-3	4
11-15	13.3	%VM	T	84177	1,6-0-3	3
11-15	13.8	%VM	T	84184	1,6-0-3	
11-15	12.9	%VM	T	84184 84191	1,6-0-3	4 4
11-15	13.1	%VM				43
	****	%VM			•	
11-15	****	%VM				4 *
	****	%VM				
11-15	****	%VM				4
11-15 11-15 11-15	****					3
11-15 11-15 11-15 11-15	****	%VM				5
11-15 11-15 11-15 11-15 11-15	****					7
11-15 11-15 11-15 11-15 11-15 11-15 11-15						8 9
	11-15 11-15 11-15 11-15 11-15 11-15 11-15 11-15	11-15 **** 11-15 **** 11-15 **** 11-15 **** 11-15 **** 11-15 **** 11-15 **** 11-15 **** 11-15 **** 11-15 **** 11-15 **** 11-15 **** 11-15 **** 11-15 **** 11-15 ****	11-15 **** %VM 11-15 **** %VM	11-15 **** %VM I 11-15 **** %VM T 11-15 **** %VM T	11-15 **** %VM 1 84198 11-15 **** %VM T 84205 11-15 **** %VM T 84212 11-15 **** %VM T 84219 11-15 **** %VM T 84226 11-15 **** %VM T 84226 11-15 **** %VM T 83192 11-15 **** %VM T 83200 11-15 13.1 %VM T 83206	11-15 **** %VM I 84198 1,6-0-3 11-15 **** %VM T 84205 1,6-0-3 11-15 **** %VM T 84212 1,6-0-3 11-15 **** %VM T 84219 1,6-0-3 11-15 **** %VM T 84219 1,6-0-3 11-15 **** %VM T 84226 1,6-0-3 11-15 **** %VM T 83192 2,7-1.5-5 11-15 **** %VM T 83200 2,7-1.5-5 11-15 **** %VM T 83200 2,7-1.5-5 11-15 13.1 %VM T 83206 2,7-1.5-5

Table B-1. Continued.

Гуре	Date	Sample#	Reading	Units	Туре	Date	Sample#	Reading	Unit
Г	83220	2,7-1.5-5	8	cbar	т	83305	3,8-5-3	15	cbar
Г	83227	2,7-1.5-5	9	cbar	Т	83311	3,8-5-3	15	cbar
Г	83241	2,7-1.5-5	6	cbar	Т	83318	3,8-5-3	15	cbar
Г	83250	2,7-1.5-5	6	cbar	Т	83325	3,8-5-3	16	cbar
Г	83255	2,7-1.5-5	7	cbar	Т	83332	3,8-5-3	0	cbar
r	83262	2,7-1.5-5	8	cbar	Т	83339	3,8-5-3	*	cbar
Г	83271	2,7-1.5-5	8	cbar	T	83346	3,8-5-3	*	cbar
Г	83283	2,7-1.5-5	8	cbar	T	83353	3,8-5-3	*	cbar
Г	83297	2,7-1.5-5	8	cbar	Ť	83361	3,8-5-3	*	cbar
T	83305	2,7-1.5-5	9	cbar	T	84003	3,8-5-3	*	cbar
Г	83311	2,7-1.5-5	9	cbar	T	84009	3,8-5-3	*	cbar
Т	83318	2,7-1.5-5	8	cbar	T	84016	3,8-5-3	*	cbar
T	83325	2,7-1.5-5	0	cbar	T	84030	3,8-5-3	*	cbar
T T	83332	2,7-1.5-5	0	cbar	T	84037	3,8-5-3	*	cbar
			*	cbar	T	84037	3,8-5-3	0	cbar
T	83339	2,7-1.5-5	*		T	84044 84052		*	
T	83346	2,7-1.5-5	*	cbar	(3,8-5-3		cbar
Г	83353	2,7-1.5-5	*	cbar	T	84058	3,8-5-3	0	cbar
Т	83361	2,7-1.5-5	*	cbar	T	84065	3,8-5-3	0	cbar
Г	84003	2,7-1.5-5		cbar	T	84072	3,8-5-3	0	cbar
Т	84009	2,7-1.5-5	*	cbar	T	84079	3,8-5-3	0	cbar
Т	84016	2,7-1.5-5	*	cbar	Т	84086	3,8-5-3	0	cbar
Т	84030	2,7-1.5-5	*	cbar	Т	84093	3,8-5-3	0	cbar
Т	84037	2,7-1.5-5	*	cbar	Т	84100	3,8-5-3	0	cbar
Г	84044	2,7-1.5-5	0	cbar	Т	84107	3,8-5-3	1	cbar
Т	84052	2,7-1.5-5	*	cbar	Т	84114	3,8-5-3	1	cbar
Г	84058	2,7-1.5-5	*	cbar	Т	84121	3,8-5-3	1	cbar
r	84065	2,7-1.5-5	*	cbar	Т	84128	3,8-5-3	2	cbar
Г	84072	2,7-1.5-5	0	cbar	Т	84135	3,8-5-3	3	cbar
Г	84079	2,7-1.5-5	0	cbar	Т	84142	3,8-5-3	3	cbar
г	84086	2,7-1.5-5	1	cbar	Т	84151	3,8-5-3	6	cbar
Т	84093	2,7-1.5-5	0	cbar	Т	84156	3,8-5-3	6	cbar
Т	84100	2,7-1.5-5	0	cbar	Т	84163	3,8-5-3	6	cbar
Т	84107	2,7-1.5-5	0	cbar	Т	84170	3,8-5-3	15	cbar
Т	84114	2,7-1.5-5	0	cbar	Т	84177	3,8-5-3	19	cbar
T	84121	2,7-1.5-5	0	cbar	Т	84184	3,8-5-3	20	cbar
T	84128	2,7-1.5-5	0	cbar	T	84191	3,8-5-3	15	cbar
Г Т	84135	2,7-1.5-5	0	cbar	T	84198	3,8-5-3	17	cbar
T	84142	2,7-1.5-5	ů 0	cbar	T	84205	3,8-5-3	20	cbar
T	84151	2,7-1.5-5	Õ	cbar	Т	84212	3,8-5-3	**	cbar
T	84156	2,7-1.5-5	0	cbar	T	84219	3,8-5-3	8	cbar
T		2,7-1.5-5	0	cbar	T	84226	3,8-5-3	8	cbar
I T	84163	,	5	cbar	T	83192	3,8-5-3 4,9-6-5	8 16	cbar
	84170	2,7-1.5-5							
r T	84177	2,7-1.5-5	5	cbar	T	83200	4,9-6-5	15	cbar
Т	84184	2,7-1.5-5	5	cbar	T	83206	4,9-6-5	19	cbar
T	84191	2,7-1.5-5	4	cbar	T	83213	4,9-6-5	20	cbar
Г	84198	2,7-1.5-5	4	cbar	Т	83220	4,9-6-5	20	cbar
Г	84205	2,7-1.5-5	3	cbar	Т	83227	4,9-6-5	20	cbar
Г	84212	2,7-1.5-5	*	cbar	Т	83241	4,9-6-5	13	cbar
Т	84219	2,7-1.5-5	3	cbar	Т	83250	4,9-6-5	11	cbar
Т	84226	2,7-1.5-5	4	cbar	Т	83255	4,9-6-5	12	cbar
Г	83200	3,8-5-3	22	cbar	Т	83262	4,9-6-5	14	cbar
Г	83206	3,8-5-3	24	cbar	Т	83271	4,9-6-5	15	cbar
Г	83213	3,8-5-3	29	cbar	Т	83283	4,9-6-5	15	cbar
Г	83220	3,8-5-3	28	cbar	Т	83297	4,9-6-5	15	cbar
Г	83227	3,8-5-3	27	cbar	Т	83305	4,9-6-5	15	cbar
Г	83241	3,8-5-3	13	cbar	T	83311	4,9-6-5	16	cbar
Г	83250	3,8-5-3	13	cbar	T	83318	4,9-6-5	16	cbar
Г	83255	3,8-5-3	15	cbar	T	83325	4,9-6-5	17	cbar
T	83262	3,8-5-3	16	cbar	T	83332	4,9-6-5	0	cbar
Г Г	83271	3,8-5-3	16	cbar	T	83339	4,9-6-5	*	cbar
	83283	3,8-5-3 3,8-5-3	16	cbar	T	83346	4,9-6-5	*	cbar
Т									

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Table B-1. Continued.

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Туре	Date	Sample#	Reading	Units	Туре	Date	Sample#	Reading	Units
Т	83361	4,9-6-5	*	cbar	т	83283	5,10-11-10	11	cbar
Т	84003	4,9-6-5	*	cbar	Т	83297	5,10-11-10	10	cbar
Г	84009	4,9-6-5	*	cbar	Т	83305	5,10-11-10	12	cbar
Т	84016	4,9-6-5	*	cbar	Т	83311	5,10-11-10	13	cbar
Г	84030	4,9-6-5	*	cbar	Т	83318	5,10-11-10	13	cbar
Т	84037	4,9-6-5	*	cbar	Т	83325	5,10-11-10	14	cbar
Г	84044	4,9-6-5	4	cbar	Т	83332	5,10-11-10	0	cbar
Т	84052	4,9-6-5	4	cbar	Т	83339	5,10-11-10	*	cbar
Т	84058	4,9-6-5	5	cbar	Т	83346	5,10-11-10	*	cbar
Т	84065	4,9-6-5	6	cbar	Т	83353	5,10-11-10	*	cbar
Т	84072	4,9-6-5	7	cbar	Т	83361	5,10-11-10	*	cbar
Т	84079	4,9-6-5	8	cbar	Т	84003	5,10-11-10	*	cbar
т	84086	4,9-6-5	11	cbar	Т	84009	5,10-11-10	*	cbar
Т	84093	4,9-6-5	12	cbar	Т	84016	5,10-11-10	*	cbar
Т	84100	4,9-6-5	14	cbar	Т	84030	5,10-11-10	*	cbar
Т	84107	4,9-6-5	14	cbar	Т	84037	5,10-11-10	*	cbar
Т	84114	4,9-6-5	9	cbar	Т	84044	5,10-11-10	0	cbar
Г	84121	4,9-6-5	16	cbar	Т	84052	5,10-11 - 10	0	cbar
Г	84128	4,9-6-5	15	cbar	Т	84058	5,10-11-10	0	cbar
Г	84135	4,9-6-5	16	cbar	Т	84065	5,10-11-10	0	cbar
Г	84142	4,9-6-5	14	cbar	Т	84072	5,10-11-10	0	cbar
Т	84151	4,9-6-5	16	cbar	Т	84079	5,10-11-10	1	cbar
Г	84156	4,9+6-5	16	cbar	Т	84086	5,10-11-10	2	cbar
Т	84163	4,9-6-5	16	cbar	Т	84093	5,10-11-10	2	cbar
Т	84170	4,9-6-5	15	cbar	Т	84100	5,10-11-10	2	cbar
Т	84177	4,9-6-5	15	cbar	Т	84107	5,10-11-10	2	cbar
Т	84184	4,9-6-5	13	cbar	Т	84114	5,10-11-10	1	cbar
Т	84191	4,9-6-5	14	cbar	Т	84121	5,10-11-10	3	cbar
г	84198	4,9-6-5	16	cbar	Т	84128	5,10-11-10	4	cbar
Т	84205	4,9-6-5	19	cbar	Т	84135	5,10-11-10	4	cbar
Г	84212	4,9-6-5	**	cbar	Т	84142	5,10-11-10	3	cbar
Г	84219	4,9-6-5	15	cbar	Т	84151	5,10-11-10	5	cbar
Г	84226	4,9-6-5	12	cbar	Т	84156	5,10-11-10	4	cbar
Г	83192	5,10-11-10	6	cbar	Т	84163	5,10-11-10	4	cbar
Г	83200	5,10-11-10	6	cbar	Т	84170	5,10-11-10	4	cbar
Г	83206	5,10-11-10	5	cbar	Т	84177	5,10-11-10	4	cbar
r	83213	5,10-11-10	11	cbar	Т	84184	5,10-11-10	2	cbar
Г	83220	5,10-11-10	8	cbar	Т	84191	5,10-11-10	4	cbar
Г	83227	5,10-11-10	11	cbar	Т	84198	5,10-11-10	3	cbar
Г	83241	5,10-11-10	6	cbar	Т	84205	5,10-11-10	1	cbar
Г	83250	5,10-11-10	6	cbar	Т	84212	5,10-11-10	*	cbar
Г	83255	5,10-11-10	9	cbar	Т	84219	5,10-11-10	2	cbar
Г	83262	5,10-11-10	10	cbar	Т	84226	5,10-11-10	2	cbar
Т	83271	5,10-11-10	12	cbar)	··· · · · · · ·			

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