Hydrology/Radionuclide Migration Program and Related Research Activities

FY 1986 Progress Report (October 1, 1985 – September 30, 1986)

Compiled by

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JUL 23 1982

Prepared for

Nevada Field Office
U.S. Department of Energy
Under Contract No. DE-AC08-85NV10384

February 1992

MASTER

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INTRODUCTION

This report presents the results of technical studies conducted under the Hydrology/Radionuclide Migration Program (HRMP) at the Nevada Test Site (NTS) for the period of October 1, 1985 through September 30, 1986.

The HRMP was initiated in 1973 as the Radionuclide Migration Program to study and better understand the hydrologic systems of the NTS and potential movement and rates of movement of radionuclides and other contaminants injected into these systems by underground nuclear testing. It began, and continues, as a multi-agency research project. The cooperating agencies are: 1) Desert Research Institute (DRI), University of Nevada System; 2) Lawrence Livermore National Laboratory (LLNL); 3) Los Alamos National Laboratory; 4) U.S. Geological Survey (USGS); and 5) Nevada Field Office of the U.S. Department of Energy (DOE/NV), which provides guidance and funding.

The original intent of this document was to reorganize all four agencies existing annual reports for this reporting period and place them under one cover. However, due to the varying typesets and formats of the individual reports, it became evident that this would be a very time consuming and difficult task. Therefore, the individuality of the four reports has been maintained in this document.

The following is a brief summary of the research activities of the HRMP for FY 1986.

Desert Research Institute

The DRI's efforts for FY 1986 consisted of three major endeavors. Section 1 describes the investigation into the effects of surface collapse structures on infiltration and moisture redistribution, in an attempt to evaluate the effects of man-made surficial features (subsidence craters) on groundwater recharge at the NTS.

Section 2 summarizes an investigation of the spatial variability of hydraulic properties in undisturbed alluvial soil. The general goals were to work toward an equation set describing the phenomena known to occur subsurface but not accounted for in the equation sets on which current numerical models are based. The ultimate result of this work will be a methodology for data gathering and numerical modeling that is physically based enough to allow actual predictions of the movement of heat, water, vapor and impurities in the vadose zone at the NTS.

Section 3 describes an ongoing effort to develop a three-dimensional model of tritium and chlorine-36 from the Cambrian cavity to a nearby pumping well (RNM-2s). The objectives of this investigation were to study the transport process and verify existing theories of contaminant transport and radionuclide solubility by inducing rapid migration of nuclides to the monitoring point.

Lawrence Livermore National Laboratory

LLNL's report is organized on a topical basis. Section 1 of the original LLNL report of FY 1985 and FY 1986 HRMP activities (UCRL-53779) is a general introduction, which has been omitted here. Section 2 summarizes the current investigations into colloid trans-

port of radionuclides at the Cheshire site and includes a review of the literature concerning colloid transport in general.

Section 3 describes experiments conducted at the Cambric site where flow velocity in a seepage plume from an unlined ditch carrying water pumped from the Cambric cavity has been characterized. Water metabolism was studied in plants growing along the unlined ditch by making use of tritium in the water.

Section 4 describes a method developed for ^{99}Tc analysis of water samples and the results of analysis of samples from both present and historical HRMP sites.

Section 5 describes several new measurements on samples collected in NTS Area 2 in 1975.

The report concludes with a description of several methodological investigations relating to analytical techniques and quality assurance, and two appendices showing various data.

Los Alamos National Laboratory

LANL continued to monitor the transport of tritium and ^{85}Kr from the Cambric explosion zone to the satellite well, which is pumped at 600 gal/min. Corresponding movement of cationic radionuclides such as ^{137}Cs and ^{90}Sr has not yet been observed after 12 years of pumping, nor has there been evidence that these strongly sorbing ions move in conjunction with colloids. More data from the Cheshire site were analyzed, but the uncertainties have not been resolved regarding the distribution and movement of radioactive materials at this location. Attempts to improve the analytical capability for ^{36}Cl and ^{99}Tc have resulted in some progress. Similarly, a better understanding has been achieved of radionuclide transport phenomena such as channeling in fracture flow and anion exclusion in zeolites and clays. A sample exchange with LLNL has helped in identifying critical steps in the procedures for collecting and analyzing large-volume water samples. Potential sites on Pahute Mesa at the NTS have been surveyed for future radionuclide migration studies and it has been concluded that there are none other than Cheshire presently available, and none are likely to be created in the near future. LANL has engaged recently in radionuclide migration studies sponsored by the weapons program, and a review of this work appears in an appendix to this report.

U.S. Geological Survey

RNM Program activities conducted by the USGS consisted of collecting and processing hydrologic data and entering these data into the USGS/Water Resources Division computerized groundwater data base system, managed by the Nevada State Office in Carson City.

Groundwater levels were monitored continuously at three wells and intermittently at selected wells, test holes, and emplacement holes. Selected measurements for the Nash and Bourbon satellite wells and the Cheshire well are listed, and changes in water levels measured at these wells are described.

The data collection and analysis effort on the Surface Disruption Study, a program designed to determine the significance of surface subsidence craters created by underground nuclear detonations to groundwater recharge, is described.

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1. EFFECTS OF SURFACE COLLAPSE STRUCTURES ON INFILTRATION AND MOISTURE REDISTRIBUTION

INTRODUCTION

In an attempt to evaluate the effects of man-made surficial features (subsidence craters) on groundwater recharge at the Nevada Test Site (NTS), we have been conducting studies at subsidence crater U3fd since 1980. Researchers have long suspected that natural features such as ephemeral stream channels and fractured bedrock might act as conduits through which atmospheric precipitation could easily infiltrate to the saturated zone. Similarly, it was thought that over 400 craters at the NTS might serve as point sources of recharge, concentrating surface-water runoff above the rubble chimneys which, in turn, might provide hydraulic connection through the unsaturated valley-fill sediments to the water table. Rather than addressing the nature of fluid transport through the rubble chimney, this investigation dealt specifically with moisture movement in the shallow regions of the unsaturated zone below the crater.

Deep (>5 meter) monitoring of the soil profile was necessary to determine if downward movement of water was occurring as a result of the surface features. We completed two boreholes and collected core data on the in-situ moisture conditions and access for neutron moisture logging tools in both disturbed and undisturbed areas. This report outlines the results of that drilling.

BOREHOLE CONSTRUCTION

Borehole U3fd-N1 was drilled to a depth of 46.4 meters within subsidence crater U3fd in an attempt to penetrate through the zone of active infiltration and into sediments whose moisture conditions were unaffected by the surface disruption. We drilled the second borehole, U3fd-N2, to a depth of 30.7 meters in nearby sediments which appeared to be unaffected by weapons testing. We had hoped that N2 would provide data on the undisturbed moisture conditions representative of a deep, unsaturated zone with little or no active recharge.

Our sample collection consisted of periodic 60 cm drive cores supplemented with cutting samples collected every 1.5 meters. Drive cores provided data on in-situ moisture conditions. Although unsuitable for representative moisture or density determinations because of drying, these cutting samples could be used for tritium and other chemical analyses.

LABORATORY RESULTS OF CORE SAMPLES AND DRILL CUTTINGS

Core analysis from borehole N1 indicated high moisture contents and very low capillary pressures (>-0.1 bar). A unit hydraulic gradient was observed throughout the

borehole. Using core data, a downward flux of at least 5.4×10^{-1} m/year and a minimum average pore water velocity of 4.4 m/year was calculated. In the undisturbed area, core data showed much lower moisture contents and matric potentials of between -8 and -36 bars. Matric potentials are shown for both boreholes in Figure 1. Gradients calculated in the undisturbed zone were erratic with depth, indicating both upward and downward flow. These conditions are typical of soils receiving little or no recharge (Gee and Heller, 1985). No soil moisture flux estimates were made for the core data in the undisturbed environment due to the conflicting gradient measurements.

Calibrated neutron logs were run on both boreholes and indicated a significantly higher stored moisture content in the soils below the crater, further indicating the theory of enhanced recharge in the crater environment. In the undisturbed borehole, 50 percent lower moisture storage was observed as well as much lower variability in moisture content.

Although simplified methods were used to determine the infiltration below the crater environment, it is clear that this environment significantly increased the recharge potential. Data from the undisturbed soils showed little or no detectable soil water movement.

By assuming that the infiltrating water moves vertically downward below the crater bottom, the velocity of the wetting front is at least 4.4 m/year. By using an average precipitation value of 0.168 m/year, a minimum of 13 percent of the precipitation falling on the crater catchment results in deep percolation.

CONCLUSIONS

Several in-situ experiments have been used to estimate the magnitude of soil-moisture movement in the alluvial-filled valleys of the NTS and surrounding areas. In all but one case, soil-moisture movement was very slow with fluxes and velocities less than several centimeters per year. In only one area, where precipitation has been concentrated and ponded due to weapons testing (subsidence craters), was significant downward movement detected. Measured matric potentials were less than -0.1 bar with most considerably lower. Field-moisture content ranged from 4 to 17 percent by weight in the undisturbed area, while the average volumetric-moisture content at the active flow site (subsidence crater U3fd) ranged from 20 to 37 percent. It is clear from this data, therefore, that the crater environment of Yucca Flat is likely to significantly increase or enhance the groundwater recharge in the valley.

FIGURE 1. Capillary Pressure Profile from Cores taken from U3fd-N1 and U3fd-N2.

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2. SPATIAL VARIABILITY OF HYDRAULIC PROPERTIES IN AN UNDISTURBED ALLUVIAL SOIL

INTRODUCTION

In the early 1900's, as relationships between soil characteristics and crop growth were realized, scientists became interested in quantifying the variability of various soil properties. Studies which endeavored to describe the variability of soil hydraulic properties used the conventional statistical approach of treating the observations as being statistically independent regardless of spatial position (Nielsen et al., 1973; Biggar and Nielsen, 1976; Carvello et al., 1976, Bakr, 1978; Cameron, 1978). This approach does not account for or attempt to describe the variability in hydraulic properties caused by differences in sample location. Many believe that a more complete approach to the treatment of field variability should consider the positions within the study area where observations were taken and thus, the separation distance between measurements. One method of illustrating variability as a function of separation distance between observations is the semivariogram.

A semivariogram expresses changes in a property over distance and the degree to which nearby observations depend on each other by comparing the (spatial) series of measurements to itself. Information obtained from such applications of the semivariogram can be used to create accurate maps of soil properties by using a method called kriging. Kriging is an interpolation technique based upon the semivariogram which produces estimates of values in unsampled areas without bias and with minimum and known variance.

This study presents statistical analyses of the variabilities of parameters related to unsaturated hydraulic conductivity using an infiltration method which requires that only moisture content be monitored during redistribution. These parameters include steady-state, field-saturated hydraulic conductivity, K_0 , steady-state, field-saturated moisture content, θ_0 , and a proportionality coefficient, β , where β is a parameter needed to estimate the relationship of hydraulic conductivity to volumetric water content. Spatial variability, and variability with depth, were investigated. Spatial distributions of these parameters are displayed using kriged contour maps. The possibility of relationships between observed variabilities and the soil caused by depositional processes and/or the presence of soil horizons was evaluated.

FIELD METHODS

Individual sampling sites were constructed in such a way that water could be ponded and the changes in moisture content below the pond monitored through time. Libardi et al. (1980) describe a field technique for determining steady-state hydraulic conductivity values, K_0 , which requires only that volumetric moisture content, θ , be

monitored through time. The impetus for their research was to devise a simple infiltration method of estimating $K(\theta)$ relationships without using tensiometers and complex mathematical curve-fitting techniques. The method was chosen because it had recently been shown by Jones and Wagenet (1984) to produce data in a minimal amount of time. The method also yields in-situ values from undisturbed soil at numerous depths.

Samples were taken in two sets of two transects each in areas close to the Area 5 radioactive-waste management site. Transects A and C comprised one set and are perpendicular to the local elevation contours. Transects B and D were parallel to the local elevation contours. This arrangement allowed statements to be made concerning the relationship between the spatial structure of measured properties and the depositional environment. The minimum spacing between infiltration plots was 7.5 meters.

The soil hydraulic properties, K_0 , θ_0 , and β , were measured at depths of 30, 61, and 91 cm at each of 40 sampling sites located within the study area of undisturbed soil.

Using traditional chi-squared (χ^2) analysis, K_0 was found to be lognormally distributed at all three depths, while θ_0 was found to be normally distributed at a level of significance of 2.5 percent at all three depths. The dimensionless parameter, β , was found to be normally distributed at the same level of significance for 30 cm and 61 cm and lognormally distributed at 91 cm.

After visual inspection of the total experimental semivariograms, only three data sets indicated possible structure. These data at 30 cm were $\log(K_0)$ and θ_0 , and at the 61 cm depth were values of $\log(K_0)$. Figure 1 shows one example of the correlation structure. The remaining data sets were found to exhibit pure nugget effects through the range of the transects.

Directional semivariograms were calculated using ranges of directions and separation distances. This resulted in one semivariogram for directions perpendicular to elevation contours and one for directions parallel to contours.

Of the data analyzed, only the directional semivariograms for $\log(K_0)$ at 30 cm revealed significant differences that may be attributed to anisotropy. These are shown in Figure 2. The anisotropy could be due to highly directional processes, such as stream deposition of detritus, which affect the fabric of the alluvial deposit and, thus, the soil which develops on it. The periodicity of the semivariogram for directions parallel to elevation contours may reflect the periodic nature of the braided channels in this direction. If these channels are responsible for giving a particular fabric to the

FIGURE 1. Total Semivariograms for $\log K_o$ at 30 cm. Best Fit Theoretical Model is Shown.

FIGURE 2. Directional Semivariograms for $\log K_o$ at 30 cm. Best Fit Theoretical Model is Shown.

soil at any particular time, then they may also be responsible for giving variance structure to hydraulic properties.

Contour maps of the three parameters shown to have variance structures ($\log K_0$ at 30 cm, θ_0 at 30 cm, and $\log K_0$ at 61 cm) were constructed using kriging techniques. Figure 3 shows the kriged estimate of $\log K_0$ at the 30 cm depth as well as the contour map of the kriging standard error.

The fact that some of the hydraulic properties' variation was structured in the horizontal plane implied two things. First, it was reasonable to suggest that the depositional environment may, to some extent, have affected how hydraulic properties were distributed in space. The results of this study showed that sampling conducted in areas where depositional processes are known should be structured to account for the directional aspects of the processes. More specifically, samples should be taken parallel to the dominant direction of deposition and perpendicular to it. In this way, possible anisotropy may be distinguished. The second implication was that the sampling method used in this study was sensitive enough to permit structure variation of the hydraulic properties to be observed.

Such correlated structure must be recognized and can be used to the researcher's and engineer's advantage in the assessment of water and contaminant migration and designs of mitigative strategies in waste management.

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FIGURE 3. Kriged Contour Map (top) and Associated Standard Deviations for $\log K_0$ at 30 cm.

3. TRITIUM AND CHLORINE-36 MIGRATION FROM A NUCLEAR EXPLOSION CAVITY

INTRODUCTION

The Radionuclide Migration Project (RNM), sponsored by the DOE, was created in 1974 to determine the likelihood of radionuclides migrating offsite or to water-supply wells. The 1965 Cambric event site, on Frenchman Flat, became a major focus of this study.

Since nearly 10 years had elapsed from the time of the Cambric event to the initiation of the RNM project, it was predicted that this was sufficient time for groundwater to fill the chimney and cavity areas to the preshot static water level 73 m above the detonation point. Radioisotopic concentration in postshot debris would then be compared with groundwater concentrations. Because the Cambric cavity lay within a water-supply aquifer, contamination from the nuclear test was a concern. Other reasons for choosing the Cambric site for further study were: 1) its small scale would likely cause little change in the hydrology or geology of the immediate area; 2) being shallow and located in tuffaceous alluvium, re-entry drilling and sampling would be less costly and difficult; 3) it was not an active testing area; and 4) sufficient tritium had been deposited to conduct tracer tests.

Our objectives in the study were to develop groundwater conditions to induce rapid migration of nuclides to the monitoring point; study transport of contaminants in natural groundwater; verify existing theories of contaminant transport; and verify laboratory studies of radionuclide solubility.

We used a three-dimensional solute transport model to simulate and predict the future behavior of tritium and chlorine-36 breakthrough curve data. Figure 1 shows a vertical cross section of the cavity and critical distances and depths needed for modeling. Due to the nature of the hydrogeologic system at the RNM site, it was believed that natural flow gradients would not influence radionuclide migration from the Cambric cavity. To induce rapid migration of the nuclides generated by the 0.75 kiloton test, an artificial hydraulic gradient was created by pumping a satellite well located 91 m from the cavity. In addition to data generated from pumping the satellite well, we obtained hydrogeologic data from sidewall samples of two exploratory wells located approximately 300 m apart, and used statistical studies and other computer-simulated work to estimate the values of the hydrologic variables of the RNM site. A sensitivity study was done focusing on those variables which significantly affect the tritium and chlorine-36 breakthrough curves.

We analyzed tritium because over 99 percent of it is incorporated into the water molecule as HTO, and it is normally considered to be an ideal tracer. Tritium is also

FIGURE 1. Vertical Cross Section Showing Size of Cavity and T-Exchange Radius and Critical Distances and Depths needed for Modeling the RNM Site.

easily analyzed in water samples in very small concentrations. We analyzed chlorine-36 because breakthrough-curve data had shown that the initial arrival time and peak concentration values occurred before that of tritium.

PHYSICAL SETTING

Winograd et al. (1971) subdivided the NTS and vicinity into 10 hydrogeologic units. Four of these units are important within Frenchman Valley, where Cambric was detonated. They are, in order of decreasing age; lower-carbonate aquifer, tuff aquitard, welded-tuff aquifer, and valley-fill aquifer. The lower carbonate and welded-tuff aquifers store and transmit groundwater chiefly through secondary porosity developed along fractures. The valley-fill aquifer stores and transmits water through primary or interstitial porosity, and within this aquifer there is remarkable uniformity, with medium to coarse sand being the dominant grain size.

SUMMARY AND CONCLUSIONS

Based on the results of the numerical modeling, we made a number of significant conclusions regarding the potential for transport of radionuclides away from nuclear detonation cavities in a saturated alluvial environment:

1. The effective hydraulic conductivity of the cavity is approximately one-tenth as high as the average horizontal conductivity of the surrounding medium. Sensitivity analysis with the model shows that tritium transport away from the cavity is very sensitive to the cavity hydraulic conductivity. However, the functional relationship is that as the cavity conductivity increases, the peak concentration at the pumping well increases. The first arrival time remains approximately the same. If the average cavity conductivity is relatively high, tritium will be released in relatively high concentrations, but over a short period of time. If the cavity conductivity is low, then the tritium will be released in relatively low concentrations over a long period of time.
2. The release of tritium from the cavity is less sensitive to other parameters that were tested, such as cavity porosity, vertical hydraulic conductivity, and transverse dispersivity.
3. It was necessary to add retardation of tritium to the model in order to correctly reproduce the RNM-2s tritium breakthrough curve. The value of the distribution coefficient chosen agrees with values found in the literature, although it is not clear which physical mechanism is responsible for the retardation.
4. The arrival of chlorine-36 in advance of tritium has been previously hypothesized to be caused by anion exclusion of the chlorine-36 ion, which causes the anion to be transported faster than the average pore velocity

(advective transport). Since the tritium has been shown to be retarded, that is it arrives after the advective front, it is no longer clear whether chlorine-36 is being advanced or slightly retarded with respect to the advective front. Laboratory experiments would seem to be the only way to examine this process further.

5. The model has been compared to an additional 1000 days of tritium data since calibration (see Figure 2), and the agreement continues to be quite good. However, there is a slight divergence with the actual tritium concentrations being slightly higher than the model-predicted concentrations. Possible explanations for this discrepancy include recirculation of tritium from the RNM ditch and the possibility of a concentric sphere source term for the release of tritium from the cavity region.

A major question remaining to be answered is whether tritium is actually being delayed by adsorption, or ion exchange processes, or whether its delay can be explained by self-diffusion into immobile water regions. It will also be beneficial to determine whether chlorine-36 is experiencing accelerated movement through the system due to anion exclusion or whether it, too, is actually being delayed by the self-diffusion process to some degree. This could be tested by rewriting the governing equations to include intraclast porosity.

Strong agreement of simulated data to the field data indicates that average values within the cavity region are adequate for predictive modeling purposes.

FIGURE 2. Additional Tritium Data Compared to the Simulated Breakthrough Curve. Tritium Data are Corrected to Shot Time.

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Section 2. Radionuclide Migration at the Cheshire Event Site

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Introduction

The Cheshire Event was fired on February 14, 1976, with an announced yield in the 200- to 500-kt range. The site is on the Pahute Mesa (see Frontispiece). The device was detonated at a depth of 1167 m in a rhyolite formation; the preshot water level was at a depth of 630 m. Figure 2-1 shows the stratigraphy and vertical hydraulic gradient prior to detonation of the Cheshire device.

The Cheshire site is of interest to the Hydrology and Radionuclide Migration Program (HRMP) for three reasons:

- It is the only study site in tuff as opposed to alluvium or carbonate aquifers.
- 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.
- The site is only 8 km from the western boundary of the Nevada Test Site (NTS) in a permeable formation with a surface water table gradient trending southwest; therefore, this site is a likely candidate for relatively prompt, off-site transport of radionuclides.

Subsurface access was via a slant-drilled reentry hole that was eventually cased and perforated within the cavity. Water was intermittently pumped from the cavity (as indicated in Fig. 2-2 along with tritium activities). In May 1985, the well was plugged (as illustrated in Fig. 2-1) and reperforated in the higher-permeability zones of the formation at a position displaced about 100 m laterally and about 250 m above the upper boundary of the cavity.

Regional groundwater flows are expected to be upward (driven by the vertical hydraulic gradient) and horizontal toward the reperforated zone. For the mesa as a whole, the best estimate of transmissibility is $10^{-3} \text{ m}^2/\text{s}$ (Waddell, 1982), and a pump test near the Cheshire site

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Fig. 2-1. Vertical section of Cheshire experimental site showing sampling locations, hydrogeologic stratigraphy, and hydraulic head with depth (Blankennagel and Weir, 1973). Horizontal dimensions are not to scale.

Fig. 2-2. Pumping history and tritium activity at the Cheshire site. Tritium counting uncertainties are typically $<1\%$. Samples coded "X" are small-volume samples analyzed for ^3H only. Samples B-7 through B-10 are large-volume samples not fully processed at the time of publication. Coded symbols represent intervals when pump was off: 1) 9-9-83 to 8-1-84; 2) 10-25-84 to 4-25-85; 3) 5-14-85 to 5-23-85; 4) 7-20-85 to 9-11-85; 5) 10-1-85 to 10-8-85. (No water has been pumped since 11-7-85.)

indicated a transmissibility of $2 \times 10^{-2} \text{ m}^2/\text{s}$ in the most permeable zone. Such a range in values hinders precise estimation of transit times from the cavity to the external sampling location.

Initial studies of the radioactivity of the cavity water were reported by Buddemeier and Isherwood (1985). Included in these findings were the observations that a significant amount of the radioactivity was apparently associated with colloidal particles that passed through conventional filters.

Since many radionuclides strongly adsorb to mineral surfaces, transport with groundwater flow is expected to be limited. One of the key assumptions adopted in transport models is that the radionuclides, once adsorbed, are immobile. However, considerable laboratory and field evidence indicates that colloidal material is mobile in groundwater (McDowell-Boyer et al.,

1986), and the initial Cheshire findings revealed a possibility that radionuclides could be transported by these mobile colloids.

This section of the report presents the results of studies conducted to determine the characteristics of colloidal loading, colloidal radionuclide associations, and colloidal transport in and between the groundwater in the cavity and that in the down-gradient external formation (see Fig. 2-1). For completeness and comparison, some of the results presented by Buddemeier and Isherwood (1985) are retabulated and discussed here.

Methods

During the course of pumping from both the cavity and formation locations, water was measured in the field for pH, alkalinity, temperature, dissolved oxygen, and conductivity. Large-volume water samples were collected in new, plastic-lined, 55-gal drums, then sealed and shipped at ambient temperatures to LLNL for analysis. Storage times for the samples (before they were processed) ranged from 1 to 10 months.

In the laboratory, the contents of each drum were vigorously mixed and pumped through various filters (see Table 2-1). The first sample analyzed, B-2, was only filtered to 0.45 μm . Nuclepore polycarbonate filters were used initially for sample B-1. However, because of clogging and exceedingly slow filtration rates, we completed the sample B-1 filtration using bag-type filters (Fin-L-Filters) with nominal pore sizes of 1.0 and 0.45 μm . All the filters were then counted together and reported as 0.45 μm . For all the other samples, bag-type filters were used for 1.0-, 0.45-, and 0.20- μm nominal pore sizes. The 0.05- μm Millipore filters (293 mm in diameter) were of mixed cellulose acetate and nitrate composition; eight were used in parallel for each sample. Selected serially filtered samples were then ultrafiltered through a Millipore XX42 Pellicon cassette system with either a 100,000- or a 10,000-molecular-weight filter (nominal pore sizes of 0.006 or 0.003 μm , respectively).

For the first ultrafiltered sample, B-1, the solution was continually pumped through the filters to the ultrafilter, and the ultrafilter retentate was recycled back to the drum until the retentate volume was sufficiently reduced. Subsequent ultrafiltrations were done following a single pass through the filters into a clean drum.

The ultrafiltration system produced about 200 L of ultrafiltrate (that which passed the ultrafilter) and 0.5 L of retentate containing colloids that did not pass the ultrafilter. To determine the activity of gamma-ray—emitting radionuclides, the ultrafiltrates, retentates, and filters were evaporated to dryness, weighed, and counted for 5 days on low-background Ge(Li) detector systems. The overall uncertainties in specific activities were conservatively assumed to be $\pm 25\%$, which reflects counting statistics, calibration, and the effects of geometry (see Sec. 5 of this report). Tests showed that there was no significant loss of activity to container walls or to the ultrafilter (Buddemeier and Isherwood, 1985).

Selected retentate and ultrafiltrate samples were also analyzed by x-ray diffraction to determine the dominant mineral phases present and by x-ray fluorescence for elemental analysis. Separate analyses were undertaken for tritium and for major anions and cations. All concentrations and activities are reported per liter of original sample volume, activities are decay-corrected to the detonation time of the Cheshire Event, and detection limits are calculated at the time counted.

Details of laboratory methods have been reported previously (Buddemeier and Isherwood, 1985). See also Sec. 5 of this report for discussions on uncertainties, calibration, and specific counting methods.

Results

In this report, we shall emphasize the gamma-spectrometry and chemical results from six large-volume samples designated B-1 through B-6. Additional sample data and supporting analytical work are presented in Secs. 4 and 5 of this report. Descriptions of these samples

and the filter series we analyzed are listed in Table 2-1. Well-head measurements indicated that the cavity water had a pH of 8.6, a dissolved oxygen concentration of about 3 mg/L, and a temperature of 40 to 42°C when stabilized by protracted pumping. For the formation samples, the field pH was 8.4 with 5 to 6 mg/L dissolved oxygen; water temperature was similar to that in the cavity.

Total dissolved solids (TDS) and colloid concentrations were estimated from samples of dried ultrafiltrate and retentate. Table 2-2 summarizes these estimates and indicates the

Table 2-1. Sample descriptions.

Sample	Location	Date collected	Cumulative water pumped (L) ^a	Filters (retention size, μm) ^b
B-1	Cavity	9/8/83	2.1×10^5	1.0-0.45-0.20-0.006
B-2	Cavity	9/9/83	5×10^5	0.45
B-3	Cavity	10/23/84	1.1×10^7	1.0-0.45-0.20
B-4	Cavity	10/23/84	1.1×10^7	1.0-0.45-0.45-0.20-0.05-0.003
B-5	Formation	5/28/85	1.4×10^7 (1.04×10^6) ^c	1.0-0.45-0.20-0.05-0.003
B-6	Formation	5/28/85	1.4×10^7 (1.05×10^6) ^c	1.0-0.45-0.20-0.05-0.003

^aTotal volume pumped subsequent to installation of sampling location in cavity.

^b0.006- and 0.003- μm filters are 100,000- and 10,000-molecular-weight ultrafilters, respectively.

^cCumulative volume pumped at formation location after relocation of perforated interval and pump.

Table 2-2. Dissolved and colloidal material in Cheshire samples.

Sample	Location	TDS ^{a,b} (mg/L)	Colloid size range (μm)	Colloid mass concentration (mg/L)
B-1	Cavity	256	0.20-0.006	55
B-2	Cavity	(256)	0.45-0.006	(63)
B-3	Cavity	(263)	0.20-0.003	(35)
B-3	Cavity	(263)	0.20-0.05	(25)
B-4	Cavity	263	0.05-0.003	10.1
B-5	Formation	216	0.05-0.003	4.6
B-6	Formation	218	0.05-0.003	4.3

^aTDS = total dissolved solids.

^bParentheses indicate an assumed value based on samples collected close in time to the indicated sample.

considerable mass of submicrometer colloids present in these pumped groundwaters. The ultrafiltrates were assumed to exclude all colloids; thus, the dried ultrafiltrate is the TDS. Since ultrafiltration was not conducted for all samples, some extrapolation was necessary; this is indicated by the numbers in parentheses in Table 2-2. For example, since the TDS was not available for sample B-2, we assumed the value from B-1. Similarly, sample B-3 was collected on the same date as B-4, so for sample B-3 we assumed the TDS of 263 mg/L from sample B-4.

Dried retentate samples provided estimates of colloid mass concentration. After sample B-4 was filtered through a 0.05- μm filter, the 194.5 L was ultrafiltered, producing a retentate volume of 0.245 L with a dried mass of 2.04 g. Assuming the TDS in the retentate is the same as the ultrafiltrate provides the estimate that 10.1 mg of 0.05- to 0.003- μm colloids are present per liter of the original sample. Samples with different filtrations prior to ultrafiltration provide colloid masses in the various size ranges given in Table 2-2. It is further interesting to note that these original samples were not cloudy despite the high colloid concentrations.

Radionuclide activities retained on various filters and concentrated by ultrafiltration are compiled in Appendix A. Tables 2-3 through 2-8 present the activity data expressed as fraction of total activity of the samples B-1 through B-6, respectively. The actual order of sample processing was B-2, B-1, B-3, B-4, B-5, and B-6; the evolving procedures were dictated by the results of the analysis. For sample B-2, detection of significant activity of the low-solubility, adsorbing nuclides manganese, cobalt, cesium, and europium after the sample was passed through 0.45- μm filters suggested that this filtration did not separate dissolved from particle-associated nuclides. For sample B-3, we switched completely to bag filters (Table 2-3); again, substantial radionuclide activity passed through the 0.20- μm filter for those particle-associated nuclides. The Nuclepore filters used for sample B-2 appear to have been partially plugged because the fraction of europium passing this filter is about 80%; in comparison, about 90% of the europium activity passed through the smaller, 0.20- μm filter used for sample B-3.

Table 2-3. Cheshire nuclide analysis for cavity sample B-1.

Nuclide	Half-life (y)	Total activity ($\mu\text{Ci/mL}$) ^a	Fraction of total activity retained on				Fraction dissolved
			Prefilter	0.45 μm	0.20 μm	0.006 μm	
³ H	12.33	0.615 (1)	NA ^b	NA	NA	NA	1.000
²² Na	2.605	7.57×10^{-9} (24)	<0.001	0.061	<0.001	<0.06	0.939
⁴⁰ K	1.3×10^9	2.67×10^{-9} (18)	<0.02	<0.02	<0.02	0.625	0.375
⁵⁴ Mn	0.855	1.18×10^{-7} (25)	0.503	0.069	0.059	0.369	<0.14
⁶⁰ Co	5.272	5.53×10^{-9} (16)	0.576	0.082	0.071	0.271	<0.004
¹⁰⁶ Ru	1.020	1.25×10^{-5} (16)	0.189	0.084	0.067	0.578	0.082
¹²⁵ Sb	2.760	5.27×10^{-6} (24)	0.007	0.003	0.002	0.039	0.949
¹³⁴ Cs	2.065	2.18×10^{-8} (17)	0.055	0.020	0.016	0.322	0.587
¹³⁷ Cs	30.170	3.19×10^{-6} (16)	0.084	0.024	0.016	0.326	0.550
¹⁴⁴ Ce	0.778	1.20×10^{-8} (17)	0.280	0.074	0.055	0.591	<0.01
¹⁵² Eu	13.40	9.51×10^{-9} (17)	0.281	0.071	0.052	0.596	<0.01
¹⁵⁴ Eu	8.50	1.64×10^{-8} (17)	0.286	0.073	0.053	0.588	<0.01
¹⁵⁵ Eu	4.73	4.10×10^{-8} (16)	0.313	0.082	0.059	0.547	<0.01

^aAll activities decay-corrected to $T_0 = 14 \text{ Feb. } 1976$. Values in parentheses are the percent uncertainties ($\pm 1\sigma$) based on counting uncertainties for the individual samples.

^bNA = not analyzed.

Table 2-4. Cheshire nuclide analysis for cavity sample B-2.

Nuclide	Half-life (y)	Total activity ($\mu\text{Ci/mL}$) ^a	Fraction of total activity retained on 0.45- μm filter	Fraction of total activity <0.45 μm
³ H	12.26	0.614 (1)	NA ^b	1.000
²² Na	2.605	9.79×10^{-9} (23)	0.079	0.921
⁴⁰ K	1.3×10^9	5.24×10^{-9} (19)	0.269	0.731
⁵⁴ Mn	0.855	1.20×10^{-7} (22)	0.341	0.659
⁶⁰ Co	5.272	6.07×10^{-9} (18)	0.332	0.668
¹⁰⁶ Ru	1.020	1.48×10^{-5} (21)	0.185	0.815
¹²⁵ Sb	2.760	6.27×10^{-6} (25)	0.007	0.993
¹³⁴ Cs	2.065	2.68×10^{-8} (21)	0.182	0.818
¹³⁷ Cs	30.170	3.40×10^{-6} (20)	0.212	0.788
¹⁴⁴ Ce	0.778	1.49×10^{-8} (20)	0.209	0.791
¹⁵² Eu	13.40	1.20×10^{-8} (21)	0.197	0.803
¹⁵⁴ Eu	8.50	1.90×10^{-8} (21)	0.169	0.831
¹⁵⁵ Eu	4.73	6.10×10^{-8} (21)	0.191	0.809

^aAll activities decay corrected to $T_0 = 14 \text{ Feb. } 1976$. Values in parentheses are the percent uncertainties ($\pm 1\sigma$) based on counting uncertainties for the individual samples.

^bNA = not analyzed.

Table 2-5. Cheshire nuclide analysis for cavity sample B-3.

Nuclide	Half-life (y)	Total activity ($\mu\text{Ci/mL}$) ^a	Fraction of total activity retained on			Fraction of total activity <0.20 μm
			Prefilter	0.45 μm	0.20 μm	
³ H	12.26	0.504 (1)	NA ^b	NA	NA	1.000
²² Na	2.605	8.02×10^{-9} (25)	0.019	<0.008	<0.002	0.981
⁴⁰ K	1.3×10^9	3.58×10^{-9} (24)	[Combined filters, 0.03]			0.097
⁵⁴ Mn	0.855	9.64×10^{-8} (64)	[Combined filters, 0.128]			0.872
⁶⁰ Co	5.272	3.42×10^{-9} (22)	0.068	0.035	0.031	0.866
¹⁰⁶ Ru	1.020	1.14×10^{-5} (23)	0.029	0.029	0.025	0.917
¹²⁵ Sb	2.760	6.77×10^{-6} (25)	0.001	0.002	0.001	0.996
¹³⁴ Cs	2.065	2.70×10^{-8} (37)	0.008	ND ^b	ND	0.992
¹³⁷ Cs	30.170	4.60×10^{-6} (24)	0.009	0.007	0.008	0.976
¹⁴⁴ Ce	0.778	1.01×10^{-8} (24)	0.051	<0.004	<0.003	0.949
¹⁵² Eu	13.40	7.52×10^{-9} (23)	0.047	0.024	0.027	0.902
¹⁵⁴ Eu	8.50	1.31×10^{-8} (23)	0.044	0.025	0.026	0.905
¹⁵⁵ Eu	4.73	3.87×10^{-8} (23)	0.044	0.022	0.023	0.911

^aAll activities decay-corrected to $T_0 = 14 \text{ Feb. } 1976$. Values in parentheses are the percent uncertainties ($\pm 1\sigma$) based on counting uncertainties for the individual samples.

^bNA = not analyzed; ND = not detected; no limit calculated.

Table 2-6. Cheshire nuclide analysis for cavity sample B-4.

Nuclide	Half-life (y)	Total activity ($\mu\text{Ci/mL}$) ^a	Fraction of total activity retained on					Fraction dissolved
			Prefilter	0.45 μm	0.20 μm	0.05 μm	0.003 μm	
³ H	12.36	0.504 (1)	NA ^b	NA	NA	NA	NA	1.000
²² Na	2.605	8.5×10^{-9} (23)	<0.18	<0.01	0.022	0.013	0.037	0.928
⁴⁰ K	1.3×10^9	4.15×10^{-9} (20)	0.198	0.039	0.079	0.330	0.084	0.270
⁵⁴ Mn	0.855	3.31×10^{-8} (36)	0.750	0.250	<0.14	<0.12	ND ^b	ND
⁶⁰ Co	5.272	3.12×10^{-9}	0.368	0.129	0.155	0.123	0.204	0.021
¹⁰⁶ Ru	1.020	1.05×10^{-6} (22)	0.321	0.129	0.158	0.070	0.178	0.144
¹²⁵ Sb	2.760	7.30×10^{-6} (24)	0.006	0.005	0.004	0.003	0.014	0.968
¹³⁴ Cs	2.065	2.67×10^{-8} (19)	0.111	0.038	0.048	0.067	0.057	0.679
¹³⁷ Cs	30.170	4.39×10^{-6} (19)	0.116	0.041	0.055	0.068	0.075	0.645
¹⁴⁴ Ce	0.778	9.56×10^{-9} (25)	0.362	0.132	0.140	0.071	0.295	<0.05
¹⁵² Eu	13.40	6.75×10^{-9} (25)	0.359	0.127	0.162	0.135	0.209	0.008
¹⁵⁴ Eu	8.50	1.17×10^{-8} (25)	0.364	0.130	0.161	0.127	0.218	<0.01
¹⁵⁵ Eu	4.73	3.45×10^{-8} (25)	0.345	0.118	0.148	0.109	0.280	<0.03

^aAll activities decay-corrected to $T_0 = 14$ Feb. 1976. Values in parentheses are the percent uncertainties ($\pm 1\sigma$) based on counting uncertainties for the individual samples.

^bNA = not analyzed; ND = not detected; no limit calculated.

Table 2-7. Cheshire nuclide analysis for formation sample B-5.

Nuclide	Half-life (y)	Total activity ($\mu\text{Ci/mL}$) ^a	Fraction of total activity retained on					Fraction dissolved
			Prefilter	0.45 μm	0.20 μm	0.05 μm	0.003 μm	
³ H	12.36	0.433 (1)	NA ^b	NA	NA	NA	NA	1.000
²² Na	2.605	1.07×10^{-9} (24)	<0.02	<0.02	0.057	<0.28	<0.20	0.943
⁴⁰ K	1.3×10^9	2.94×10^{-9} (20)	0.047	<0.01	<0.01	0.118	0.083	0.752
⁵⁴ Mn	0.855	7×10^{-9} (72)	[Combined filters, 1.0]					<0.01
⁶⁰ Co	5.272	1.06×10^{-10} (25)	0.441	<0.05	0.097	0.294	0.168	<0.04
¹⁰⁶ Ru	1.020	2.67×10^{-6} (20)	0.162	0.071	0.055	0.289	0.205	0.217
¹²⁵ Sb	2.760	3.93×10^{-6} (25)	0.003	0.002	0.004	0.004	0.006	0.985
¹³⁴ Cs	2.065	1.91×10^{-9} (25)	ND ^b	ND	ND	ND	0.024	0.976
¹³⁷ Cs	30.170	7.43×10^{-7} (22)	0.018	0.005	0.007	0.050	0.035	0.884
¹⁴⁴ Ce	0.778	$<7.5 \times 10^{-11}$ (—)	ND ^b	ND	ND	ND	ND	ND
¹⁵² Eu	13.40	1.65×10^{-10} (25)	0.237	<0.07	<0.08	0.458	0.305	<0.06
¹⁵⁴ Eu	8.50	2.99×10^{-10} (25)	0.245	<0.02	<0.02	0.466	0.289	<0.07
¹⁵⁵ Eu	4.73	8.8×10^{-10} (25)	0.173	0.034	0.066	0.382	0.345	<0.13

^aAll activities decay-corrected to $T_0 = 14$ Feb. 1976. Values in parentheses are the percent uncertainties ($\pm 1\sigma$) based on counting uncertainties for the individual samples.

^bNA = not analyzed; ND = not detected; no limit calculated.

Table 2-8. Cheshire nuclide analysis for formation sample B-6.

Nuclide	Half-life (y)	Total activity ($\mu\text{Ci/mL}$) ^a	Fraction of total activity retained on					Fraction dissolved
			Prefilter	0.45 μm	0.20 μm	0.05 μm	0.003 μm	
³ H	12.36	0.433 (1)	NA ^b	NA	NA	NA	NA	1.000
²² Na	2.605	1.49×10^{-9} (19)	[Combined filters, <0.01]					<0.01 >0.99
⁴⁰ K	1.3×10^9	2.79×10^{-9} (21)	0.036	<0.01	<0.01	0.094	0.020	0.850
⁵⁴ Mn	0.855	$<6 \times 10^{-9}$	ND ^b	ND	ND	ND	ND	ND
⁶⁰ Co	5.272	5.3×10^{-11} (25)	1.000	<0.01	<0.01	<0.10	<0.05	>0.09
¹⁰⁶ Ru	1.020	1.79×10^{-6} (12)	0.292	0.084	0.039	0.217	0.053	0.315
¹²⁵ Sb	2.760	3.98×10^{-6} (25)	0.003	0.001	0.001	0.002	0.002	0.991
¹³⁴ Cs	2.065	3.52×10^{-10} (25)	ND ^b	ND	ND	ND	ND	>0.90
¹³⁷ Cs	30.170	5.46×10^{-7} (23)	0.029	0.007	0.005	0.044	0.010	0.905
¹⁴⁴ Ce	0.778	$<9 \times 10^{-11}$	ND ^b	ND	ND	ND	ND	ND
¹⁵² Eu	13.40	8.98×10^{-11} (16)	0.404	<0.10	<0.10	0.492	0.104	<0.24
¹⁵⁴ Eu	8.50	1.92×10^{-10} (15)	0.453	0.120	<0.02	0.342	0.085	<0.12
¹⁵⁵ Eu	4.73	5.24×10^{-10} (16)	0.367	<0.04	<0.03	0.537	0.096	<0.39

^aAll activities decay-corrected to $T_0 = 14$ Feb. 1976. Values in parentheses are the percent uncertainties ($\pm 1\sigma$) based on counting uncertainties for the individual samples.

^bNA = not analyzed; ND = not detected; no limit calculated.

Tests of sample B-1 included the ultrafiltration step for the first time, and the analysis showed that colloidal material in the 0.20- to 0.006- μm range had a significant fraction of the radionuclides manganese, cobalt, cesium, cerium, and europium. To provide further information on radionuclide partitioning of the smaller colloids, the final analysis configuration for samples B-4 through B-6 included the 0.05- μm step. For sample B-4, two 0.45- μm filter bags were placed in series, and the second filter had about half the activity of the first. The data for the cesium and europium radioisotopes are very similar in their distribution of activity among filter sizes, and the counting statistics for a specific filter were very good. For example, in sample B-4 (Table 2-6) the fractions of total activity recorded on the 1.0- μm pre-filter were 0.111 and 0.116 for ^{134}Cs and ^{137}Cs , respectively; a similarly small variation (0.359, 0.364, and 0.345) was observed for the three europium isotopes. Comparing activity distributions between samples is complicated by the differences in samples and in counting geometry. Sample B-6 measurements had higher detection limits than sample B-5, which hindered comparison of formation samples for homogeneity.

Radionuclide transport from the cavity to the formation is dependent on physical and chemical factors. Figure 2-3 compares the ratio of formation activity to total cavity activity with the fraction dissolved for each nuclide in the cavity sample. One major effect apparent in Fig. 2-3, namely, that the tritium activity in the formation sample is 86% of that in the cavity sample, shows that very little physical dilution of the cavity water occurs.* For the cobalt and europium nuclides that are more than 98% associated with colloids, the activity at the formation location is only about 2.5% of that at the cavity—indicating significant colloid removal during solution transport. If there were no chemical exchange between dissolved nuclides and suspended colloids or between dissolved nuclides and fracture surfaces, then the other nuclides should fall in the shaded area in Fig. 2-3. Since the other nuclides are not near this mixing line, nuclide exchange among dissolved species, suspended colloids, and fracture

* A slightly greater dilution, but still less than a factor of two, is suggested by the ^{99}Tc , an anionic species often assumed to act as a conservative tracer.

Fig. 2-3. Ratio of the total radionuclide activities of formation sample B-5 to those of cavity sample B-4 as a function of dissolved fraction of total activity at the cavity location. Error bars represent cumulative counting uncertainties; data are from Table 2-6 through 2-8. Europium values are an average of the three europium isotopes measured. The shaded area represents the region in which data should fall if the nuclide transport represents a simple combination of the pure particulate (^{60}Co , ^{144}Ce , Eu) and conservative (^3H , ^{99}Tc) dissolved cases.

surfaces is suggested. ^{40}K is a special case in these results because it is a naturally occurring radionuclide; artificial production by the detonation is not believed to be significant.

The other nuclides show effects of chemical interactions during transit from the cavity to the formation location. For ^{22}Na , ^{125}Sb , ^{134}Cs , and ^{137}Cs , there is lower activity in the formation than expected if only physical dilution and colloid removal were operating, as represented by the shaded region in Fig. 2-3. This suggests either adsorption of dissolved contaminants to fracture surfaces or exchange of a radionuclide with a naturally occurring stable isotope. The result for ^{106}Ru is anomalous in that greater than expected activity is observed at the formation location.

Adsorption and desorption of nuclides can be quantified for nuclides with detectable activities in both the 0.05- to 0.003- μm range and dissolved fractions. If the linear adsorption isotherm is applicable, a distribution coefficient K_d may be expressed as

$$K_d = q/C \quad (1)$$

where q is the activity of the radionuclide adsorbed per unit area of solid, and C is the equilibrium concentration of the dissolved nuclide.

K_d values for ^{106}Ru , ^{125}Sb , ^{134}Cs , and ^{137}Cs are calculated and listed in Table 2-9 for the last cavity sample (B-4) and for the two formation samples (B-5 and B-6). Calculations were restricted to those samples because only these samples had corresponding values for colloid mass concentrations (Table 2-2). For ^{106}Ru and ^{125}Sb , the calculated distribution coefficients are close for the cavity sample and the first formation sample. The cesium radioisotopes have a greater spread in K_d values, with greater variability in ^{134}Cs than ^{137}Cs due to poorer counting statistics. For an unknown reason, formation sample B-6 was consistently low in K_d values for all the nuclides. As discussed below, K_d values calculated per mass of solids cannot be extrapolated to submicrometer colloids if they are measured on micrometer and larger particles. Cesium adsorption onto montmorillonite has a measured K_d of 4500 mL/g at 26°C in 2mM NaCl (Silva et al., 1979), a sodium concentration similar to that measured in groundwater at this site (3mM).

X-ray diffraction studies were undertaken for sample B-4 to gain some insight into the mineral composition of the colloids. The dried ultrafiltrate was composed of evaporite salts (halite, trona, aragonite), whereas the ultrafiltrate solids in the 0.05- to 0.003- μm range were dominated by quartz and (Ca,K) feldspars. About 10% of the dried ultrafilter retentate solids could not be identified by x-ray diffraction, but could be clay minerals formed as secondary minerals during feldspar weathering (Velbel, 1986). Fourier-transform, infrared spectroscopic characterizations being carried out at the Desert Research Institute (DRI) tend to support the hypothesis that clays are present in the colloid fractions (Jacobson, 1986). An equilibrium geochemical model (Wolery et al., 1984) provided further support for this interpretation by predicting chemical speciation using concentration data for 0.45- μm -filtered groundwater from sample B-2 as input.

The model predicted the following:

- Under field conditions with a pH of 8.5 and a temperature of 45°C—

Calcite	0.49 mg/L
Hausmanite	0.012 mg/L
Muscovite	8.49 mg/L
Phengite	2.57 mg/L
Quartz	49.8 mg/L

- Under laboratory conditions where the measured pH was 7.3 and the temperature was 21°C—

Kaolinite	10.1 mg/L
Nontronite-Ca	0.87 mg/L
Quartz	60.3 mg/L

Although equilibrium is not rapidly achieved in some mineral-alteration reactions, x-ray diffraction studies did observe quartz, and the existence of colloidal aluminum-silicate minerals is strongly suggested by speciation modeling.

We used x-ray fluorescence to measure the distribution of trace elements between the ultrafilter retentate and the filtrate for sample B-4. Table 2-10 lists the elements detected, the total concentration per liter of groundwater passing the 0.05- μm filters, and the fraction of that element that was present on 0.05- to 0.003- μm colloids. For potassium, manganese, iron, rubidium, cesium, barium, lanthanum, cerium, and lead, greater than 10% of each element's concentration passing the 0.05- μm filter is present on the small colloids. Detectability problems hinder a comparison of radionuclide association on colloids with elemental association, except that x-ray fluorescence indicates that 36% of the potassium passing through the 0.05- μm filter was colloidal compared to 32% of the ^{40}K detected by gamma-ray counting as passing through the 0.05- μm filter. For ^{134}Cs and ^{137}Cs , 18% and 12%, respectively, of the activity passing the 0.05- μm filters was colloidal, whereas x-ray fluorescence indicated greater than 11% colloidal. Within the uncertainties of these methods, the results are consistent and indicate that trace metals also associate with natural colloids.

Table 2-9. Calculated K_d values (mL/g) for 0.05- to 0.003- μ m-diam colloids.

Nuclide	Cavity sample	Formation samples	
	B-4	B-5	B-6
^{106}Ru	$1.1 \pm 0.4 \times 10^5$	$1.7 \pm 0.6 \times 10^5$	$0.39 \pm 0.14 \times 10^5$
^{125}Sb	$1.3 \pm 0.5 \times 10^3$	$1.3 \pm 0.5 \times 10^3$	$0.41 \pm 0.14 \times 10^3$
^{134}Cs	$7.6 \pm 3.0 \times 10^3$	$5.3 \pm 3.5 \times 10^3$	ND ^a
^{137}Cs	$10.1 \pm 4.1 \times 10^3$	$8.7 \pm 3.1 \times 10^3$	$2.6 \pm 0.9 \times 10^3$

^aND = not detected.

Table 2-10. The results of x-ray fluorescence analysis of sample B-4.

Element	Concentration passing 0.05- μ m filter (μ g/L)	Fraction colloidal; 0.05- to 0.003- μ m size range
K	735	0.36
Ca	784	0.028
Mn	31	0.18
Fe	140	0.40
Ni	8.6	0.023
Cu	2.9	0.064
Zn	530	0.011
As	13.1	0.016
Br	37	0.004
Rb	7.0	0.28
Mo	10.8	0.004
Cs	<1.5	>0.11
Ba	8.7	0.24
La	<1.5	>0.13
Ce	<2.2	>0.40
Pb	12.5	0.26

Discussion

Background

Colloids have long been recognized as potential carriers of adsorbed radionuclides in the subsurface environment (Champlin and Eicholz, 1968). The potential for transport by colloids is suggested by the colloids' submicrometer sizes, which are far smaller than pore sizes in permeable and fractured media, and by their high surface area per unit mass. Colloid removal from solution by capture onto fixed media surfaces is controlled by the Brownian motion of the colloids and by the attachment efficiency following collision. In general, for natural waters with low ionic strengths, the attachment of colloids to surfaces is hindered by electrostatic repulsion, but predictions based on double-layer theory underpredict attachment by orders of magnitude (McDowell-Boyer, 1986).

Literature on the association of radionuclides with colloids has evolved a unique nomenclature that we have not adopted here. Radionuclides such as plutonium and americium are known to form colloids with dimensions of a few nanometers (Davydov, 1967; Benes et al., 1979; Cleveland, 1979). These colloids have been called "true colloids." When radionuclides adsorb onto natural colloidal matter such as clay minerals and hydrated oxides, the resulting particles have been referred to as "pseudo-colloids" (Davydov, 1967; Benes et al., 1979). According to these definitions, a natural colloid becomes a pseudo-colloid when a radionuclide adsorbs. The designation of true versus pseudo-colloid is also inadequate in describing an aggregate composed of a radionuclide colloidal precipitate and a clay colloid. Although the chemical speciation of radionuclides with colloidal material is important, the distinction between true and pseudo-colloids is awkward for application to a broad range of problems. We recommend that the definition of colloids be based on their physical properties (i.e., that they are subject to Brownian motion and have diameters between 0.001 and 1 μm) and not on their chemical history or the scientific discipline of their observer.

The sources and properties of colloids that could transport radionuclides in natural and engineered systems are important. Apps et al. (1982) distinguish two processes for forming suspended colloids:

- Condensation or homogeneous nucleation of dissolved species when a mineral phase is supersaturated, and

- Dispersion of bulk material into suspended solids.

Dispersion of bulk materials actually comprises a number of separate processes because it can include:

- Disruption of fragile aggregates by changes in ionic strength or hydrodynamic force,
- Mechanical grinding of mineral surfaces,
- Mechanical disruption of secondary minerals present at mineral surfaces, and
- Release of less-soluble colloids by dissolution of a more-soluble matrix surrounding these colloids.

Aggregate destruction was suggested when recharge of an aquifer with water having lowered ionic strength resulted in the release of clay minerals into flowing groundwater (Nightingale and Bianchi, 1977). Similarly, infiltrating rain water causes bursts in virus concentration in well waters (Lance and Gerba, 1982). Simmons and Caruso (1983) show photographs of crystalline rocks that have been sealed by new mineral growth. Many sealed cracks have undergone repeated fracturing and sealing because the minerals in the crack are weaker than the rock and do not completely fill the void space. Similarly, Velbel (1986) presents a review of feldspar-weathering mechanisms that can include the formation of secondary minerals such as clays.

Considerable research is being conducted on the release of colloids when glasses dissolve. For example, Saltelli et al. (1984) measured the size distribution of americium-containing colloids released from a waste container, and Shade et al. (1984) measured radionuclide adsorption onto colloids released from waste-package material. Lutze et al. (1983) report on the rapid surface alteration of borosilicate glass at 165°C in distilled water. After 30 days, there was a 1- μm -thick crystalline outer layer composed of fibers with diameters $<0.1\ \mu\text{m}$.

Thus, submicrometer colloids could easily be released from mineral and glass surfaces when these surfaces are chemically, hydrodynamically, or mechanically stressed.

In spite of the existence of submicrometer colloids in natural systems, there have been few studies of contaminant adsorption to colloids of that size. Experimentally, it is far easier to use large particles that can be efficiently separated from the solution. The presence of colloids is known to interfere with the adsorption of radionuclide and organic contaminants onto micrometer and larger particles. Following conventional filtration or centrifugation, the contaminant remaining in solution is observed to increase as "nonsettleable particles" or "colloidal organic carbon" increase (Sheppard et al., 1980; Gschwend and Wu, 1985; Baker et al., 1986; Higgo and Rees, 1986). Colloidal organic carbon is assumed to be proportional to the amount of dissolved organic matter.

One aspect of the application to colloidal adsorption of adsorption measurements on micrometer and larger particles is that the linear isotherm expressed as Eq. (1) requires reevaluation. Because the isotherm relates solution activity to adsorbed activity per unit mass, experiments with smaller particles would increase the external surface area per unit mass and thus increase K_d if only the external surface area is accessible for adsorption.

James and Parks (1982) have summarized measurements of ionizable-site densities for oxide minerals; their data are spread over a relatively narrow range of 2 to 20 sites per square nanometer. Since these ionizable sites participate in adsorption reactions, relatively little variation in adsorption other than its dependence on surface-group acidity would be expected for minerals when expressed as adsorbed activity per surface area. Therefore adsorption isotherms that are measured on micrometer and larger particles and represented according to Eq. (1) should not be applied to adsorption to colloids.

Quite frequently in the literature there is reference to adsorption onto micrometer and larger particles of species that exist as colloids. Cleveland (1979) has soundly denounced such an interpretation for the interaction of plutonium oxide colloids with glass surfaces. Colloid attachment is irreversible except under aqueous solution conditions that favor dissolution of the colloid. The maximum plutonium loading on glass is $1.6 \mu\text{g}/\text{cm}^2$; if the colloids have an

average diameter of 1 nm and a density of 11.5 g/cm³, then the projected area of the colloids is twice the surface area of the glass. This loading is a physical limit of colloid packing on a surface. Cleveland has stressed that such colloid-media association cannot be described as ion-exchange phenomena; therefore, linear adsorption isotherms are completely inadequate mechanistic models. Studies of radionuclide cycling in lakes and oceans have recently acknowledged the importance of radionuclide association with colloids, and models for attachment to settling particles are being developed based on coagulation kinetics (Sanschi et al., 1986).

There are no earlier reports of field sites that demonstrate radionuclide migration by colloid transport. A number of field studies have observed anomalous, rapid, radionuclide transport that suggests mobile colloids as carriers. Travis and Nuttal (1985) have summarized the available data and reference the reports of radionuclide transport from a low-level waste site at Los Alamos National Laboratory. These reports describe how, chiefly during the period 1945–1952, liquid wastes from the plutonium purification plant were sent to adsorption beds overlaying Bandelier tuff. Core samples taken in 1978 revealed that both plutonium and americium had migrated over 30 m downward through the unsaturated tuff. Laboratory experiments on the tuff suggested that colloidal plutonium and americium were responsible for the rapid migration.

Four waste plumes at the Chalk River Nuclear Laboratories in Canada are described by Champ et al. (1984). The analytical procedures for separating gamma-emitting radionuclides distinguished only four forms: particulate species captured on 0.4- μ m filters, species retained on cation- and anion-exchange resins, and nonionic species removed by an aluminum oxide bed. For ⁶⁰Co, ⁹⁵Zr, ¹⁰⁶Ru, and ¹²⁵Sb, the anionic forms dominated, and organic ligands were felt to promote mobility for cobalt, cerium, cesium, europium, antimony, and zirconium. Even though the speciation studies would have placed colloidal species <0.4 μ m in diameter into cationic, anionic, and nonionic forms, the detection of significant particulate nuclides was

reported for ^{60}Co , ^{95}Zr , ^{106}Ru , ^{137}Cs , and ^{144}Ce . The mass of particulate matter in suspension was not reported, but since reported sampling flow rates were about 100 mL/min with 30 min of purging, the solution was probably representative of groundwater and not subject to near-well erosion.

An *in situ* glass-block leaching experiment is one of the four study sites at Chalk River. Field measurements of ^{137}Cs migration indicate transport four times farther than predicted from K_d values (Champ and Merritt, 1981). Soil columns prepared from undisturbed, uncontaminated cores showed cesium transport by 0.2- to 1.0- μm particles. There was also an indication that microorganisms were involved in particulate cesium release and transport.

Colloidal transport is also suggested in the formation of ore bodies. Horzempa and Helz (1979) have found that colloidal copper sulfide prepared by precipitation is destabilized by the ionic composition of natural waters. Transport of the colloids would then be dominated by aggregation and attachment within porous media. On the basis of laboratory experiments, Giblin et al., (1981) argued that uranium adsorbed to hydrous ferric oxide and clay colloids would be mobile in subsurface waters under certain pH and redox conditions. They noted that uranium mobility, once the uranium is adsorbed to submicrometer colloids, is controlled by colloidal stability. That is, if the colloids aggregate, limited mobility is expected.

This Study

Our results are critically dependent on filtration to separate the colloids from solution by size. We recognize a number of well-known problems with using filtration for size characteristics, but these problems do not negate the findings in this work. De Mora and Harrison (1983) have provided an overall evaluation of colloid-solution separation techniques for studies of trace-metal speciation. For depth filters used in this work, particle retention changes slowly with particle size near the stated pore size of the filter. Johnson and Wangersky (1985) have shown that removal efficiency also depends on colloid stability because particles much smaller than the size of a filter pore were removed under the high ionic strength conditions of seawater, but there was no removal when the particles were suspended in distilled water.

Another phenomenon hindering size separation by filtration is the partial clogging of filter pores as the material is retained. As a filter is loaded with particles, its effective pore size is reduced, and smaller particles are retained (Danielsson, 1982). Comparison of results obtained with etched Nuclepore filters (sample B-2) and with bag-type filters (sample B-3) indicates some clogging did occur for the Nuclepore-filtered sample.

Another issue with filtration and ultrafiltration is the intensity of fluid mixing, which could break up fragile aggregates into smaller aggregates or primary particles. Since colloid transport processes are size-dependent, any procedure that alters colloid sizes would give results that could not be used to predict transport. For groundwater samples containing colloids in dilute concentrations on the order of mg/L, there are no alternative procedures for concentrating the colloids to the extent needed for radionuclide detection.

Our experience to date with groundwater samples from the Cheshire site is that serial filtration and ultrafiltration provide consistent results that allow monitoring of radionuclide concentrations.

Deep groundwater from NTS has colloidal material that is far higher in concentration than previously reported in the literature. Apps et al. (1982) reference studies using light scattering and aluminum analysis that report colloid concentration in the $\mu\text{g/L}$ range in samples taken from unperturbed groundwater. Colloid concentrations measured in the present study are in the tens of mg/L range; these could be higher than in other areas because the fractured media of Pahute Mesa is continually subjected to blast-induced pressure pulses that could erode colloids attached to fracture-wall surfaces. Ultrafiltration can concentrate colloids larger than a few nanometers and is a more sensitive indicator of colloid concentration than methods that depend either on movements of a single element or on light scattering. The scattering effect in particular is very weak for colloids much smaller than the wavelength of the light used; therefore, this technique may tend to underestimate colloid concentrations.

Groundwater at the Cheshire site had radionuclide activity both in the cavity and in the formation. For manganese, cobalt, cerium, and europium nuclides, greater than 98% of their

activities were in the colloid fraction, and these radionuclides were detected at the formation location on submicrometer colloidal material. Less strongly sorbing nuclides such as ruthenium, cesium, and antimony are present in both colloidal and dissolved forms at both the cavity and the formation locations.

The data suggest three-phase chemical speciation:

- Dissolved nuclides,
- Nuclides associated with colloidal material with a size range of $<0.003\ \mu\text{m}$ to $1\ \mu\text{m}$, and
- Nuclides associated with immobile fracture surfaces.

Equilibrium-based predictions of contaminant migration are highly suspect in fractured media because adsorption is dependent on accessible surface area, and diffusion into the rock matrix can be very slow (Rasmuson and Neretnieks, 1986). The field data for cesium and sodium suggest some exchange of dissolved radionuclides with the fracture surfaces.

For nuclides that are nearly 100% colloidal, the total activity at the formation location is only 2.5% of the cavity activity. A comparison of colloid mass concentration over the 0.05- to $0.003\text{-}\mu\text{m}$ range shows a much smaller decrease from 10.1 mg/L at the cavity to 4.6 mg/L at the formation location. These results indicate either colloid exchange with fracture surfaces or selective removal of colloids containing radionuclides during fluid flow between the sampling locations. The process of colloid exchange with fixed surfaces is such that a mechanical, hydrodynamic, or chemical shock is required to release colloids once they are attached. Selective removal from NTS groundwater of colloids containing radionuclides is a possibility, but individual colloid mineralogy and radionuclide content are not at present experimentally accessible.

Issues

Our results raise a number of issues related to sampling, colloid characterization, colloid transport, and modeling radionuclide migration in fractured media. The drilling and well-development activities involved in collecting samples from subsurface environments are

mechanically, chemically, and hydraulically disruptive; it is not clear how these factors alter colloid generation, resuspension, and transport. At the Cheshire site, groundwater was pumped at about 30 gal/min from the >20-m-long perforated section of the reentry hole, and more than 50,000 gal were pumped before the first sample was collected. Under these conditions, the aquifer was not hydraulically stressed, and the time between initial drilling and sampling was 7 years. Since the NTS groundwater is oxygenated, sample collection and storage probably did not substantially alter the colloids or radionuclide speciation.

The colloids collected in the NTS groundwater at the Cheshire site are poorly characterized. We estimated colloid mass concentrations over broad colloidal size intervals and detected nuclide activities in these size fractions. X-ray diffraction studies indicated the major mineralogical composition, but these results are only semiquantitative. Equilibrium speciation modeling of NTS groundwater also supported the existence of mineral colloids. Obviously, better colloid characterization is necessary, but because of the dilute colloid concentration and heterogeneous distributions in size and mineralogical composition, advancements in analytical techniques are required. Also unresolved is the question of whether the high colloid loadings observed are natural or are the result of repetitive, blast-induced shocks.

Although a number of colloid-transport models for fractured media have appeared in the literature (Travis and Nuttal, 1985; Bonano and Beyeler, 1985), there is as yet much uncertainty regarding water flow, conservative tracer transport, and mechanisms of colloid generation, deposition, and erosion. Extensive model-development activities are premature without both a fundamental understanding of mechanisms and data-collection efforts in the laboratory and in the field that can be applied to model calibration and verification.

Summary

Groundwater within the Cheshire cavity contains radionuclides that are associated with submicrometer colloids. Groundwater collected outside the detonation cavity in the formation

also contained radionuclides on colloidal matter, but at reduced levels. The majority of the radionuclides associated with suspended colloids passed through conventional 0.45- and 0.20- μm filters. Chemical analysis and equilibrium speciation modeling suggest the colloidal matter is composed of secondary minerals produced by natural weathering of the fractured tuff. Although colloids were observed to transport some of the nuclides, the actual composition of colloids, the mechanisms that control their deposition and erosion, and the potential for long-distance radionuclide transport by colloids are all unknown at this time.

Section 3. Environmental Tritium Transport

Section 3A. Infiltration and Recharge (Cambric Event Site)

Contributors: M. R. Ruggieri, Scott Tyler, and R. W. Buddemeier*

Introduction

The Cambric Event site (see Frontispiece) is the location of a long-term pumping experiment designed to induce and investigate radionuclide migration in the groundwater (Hoffman, 1979; Coles and Ramspott, 1982; Hoffman and Daniels, 1984; Buddemeier and Isherwood, 1985). A result of the primary investigation has been the nearly continuous discharge, over a period in excess of 13 years, of groundwater pumped into an unlined ditch on the desert floor. This water has been continuously monitored; since late 1977 it has contained varying but well-characterized elevated levels of tritium.

In 1983, LLNL and the Desert Research Institute (DRI) cooperated in installing two aids to analysis: flumes to measure flow and infiltration rates and, adjacent to the ditch, an instrumented plot for monitoring the vadose zone. The objective was to exploit as a secondary experiment the steady-state injection of labeled water in order to study recharge and vadose-zone migration in the alluvial soils. A map of the ditch system and plan and vertical views of the vadose-zone study site are shown in Figs. 3A-1 through 3A-3. The details of the installation, sampling, and analysis are reported by Buddemeier and Isherwood (1985), as are the results through 1984.

This report updates the results of the soil-moisture and tritium-monitoring efforts and presents the moisture-transport rates calculated for the vadose zone. We also report on the installation of a deep (ca. 30-m) lysimeter immediately adjacent to the ditch and on the results of its use in a tracer study to determine directly rates of vertical migration beneath the ditch. Additional geophysical, modeling, and hydrologic characterizations have been carried out by DRI; reports on these studies are in preparation.

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Fig. 3A-1. Ditch and pond system carrying water pumped from satellite well RNM-2s. The pump is located 91 m from the working point of the Cambrian Event. The numbers in the pond correspond to sampling sites (see data in Appendix B). At the scale of this sketch, the berms along the edges of the ditch cannot be distinguished.

Fig. 3A-2. Plan view of the plume study site. The italicized numbers by the instrumentation symbols give the emplacement depth in feet.

Fig. 3A-3. Vertical view of the soil instruments and sample locations used for the vadose-zone study of the Cambric ditch. The italicized numbers give emplacement depth in feet. Contours summarizing the soil-moisture characteristics determined from tensiometer and resistance-cell data are superimposed on the plot.

Vadose-Zone Monitoring and Migration

Moisture and tritium monitoring at the vadose-zone study installation were carried out on a routine basis through late 1986; measurements were then discontinued, but the instruments remain in place, and the site potentially can be refurbished for further investigations. A complete update of the LLNL results (including the data previously published) is contained in Appendix B. LANL also monitors the tritium content of outflow to the ditch from satellite well RNM-2s; their results may be found in their annual project reports. To retain consistency with the previous data reports, the tritium data of Appendix B are reported in units of disintegrations per minute (dpm) per mL, whereas text data are presented in pCi/mL, in keeping with more recent policies to use Curie-based activity units ($1 \text{ pCi/mL} = 10^{-6} \mu\text{Ci/mL}$). Conversion factors are included in Appendix B.

The data obtained from continued monitoring support and extend the conclusions about tritium and moisture distributions presented in the previous report (Buddemeier and Isherwood, 1985). The outermost installations (3.35 m from the ditch berm; see Figs. 3A-2 and 3A-3) show no indications of elevated moisture or tritium. The soil zones described previously as wet, moist, and dry (see Fig. 3A-3) continued to exhibit those characteristics. The shallowest installations showed fluctuations in both moisture content and tritium activity that appeared to reflect natural recharge associated with precipitation events. Lysimeter A6, the closest sampling point to the ditch, continued to yield tritium activities very close to equilibrium with concurrently sampled ditch water. Lysimeter C12 continued to exhibit a pattern of tritium activity with time that was parallel to but higher than the trend in tritium activity in the ditch, as shown in Fig. 3A-4. By contrast, lysimeters D6 and E12 both showed a trend toward increasing tritium activities, suggesting that the water currently sampled at those locations originated in the ditch prior to the occurrence of the peak tritium activity in July 1980.

To determine the transport times between the ditch and the lysimeter locations, we decay-corrected tritium data to a common date (the beginning of the vadose-zone studies), and performed linear regressions of the lysimeter data against a corresponding interval of ditch

data, using a range of time lags to test for the best fit. For lysimeter C12, excellent agreement ($r^2 > 0.9$) was found for a time lag of 1.5 years; for E12 the data were somewhat noisier, but yielded a convincing transport time of 6 years. These results demonstrate the potential of the site for long-term research on the unsaturated zone.

Recharge-Plume Investigations

In May 1985, we drilled and sampled a 30.5-m borehole 2.4 m from the berm (see Fig. 3A-1) in order to sample the recharge plume directly beneath the ditch. Core samples were analyzed for tritium and water content, and a lysimeter (designated L-100 in Appendix B) was installed at 28.7 m. Samples were collected from L-100 beginning in July 1985.

Figure 3A-5 shows the sampling and installation pattern of the L-100 borehole. The water content measured in all of the cores indicated unsaturated conditions, consistent with the coarse, porous, high-permeability material. Tritium activities in the core were consistent over the length of the borehole and were close in value to the values measured in the ditch water within the preceding 1 to 2 months. The initial lysimeter samples also suggested a transport time from the ditch to the lysimeter of no more than 1 to 4 months.

Based on these preliminary results, LLNL and DRI planned and carried out a real-time experiment in which the recharge plume was traced in order to

- Compare the results with the tritium observations, and
- Conduct a feasibility study in anticipation of future large-scale tracer studies to investigate the behavior of other tracers, colloids, etc. in the recharge plume.

On March 26, 1986, a 30.5-m segment of the ditch adjacent to the deep lysimeter was dammed at both ends; large plastic tubing was used to divert flow around the blocked section. Into this isolated segment was injected 4000 L of uncontaminated (i.e., having background tritium levels) groundwater from the NTS that had been spiked with LiBr to a concentration of 237 ppm Br^- . After the labeled water had percolated into the ditch bottom, the dams were broken and normal flow was resumed. Lysimeter samples were taken at regular intervals and analyzed for Li^+ , Br^- , and tritium.

Fig. 3A-4. Comparison over time of the tritium content in the surface ditch water with that in samples from lysimeter C12 (expanded plot). The small graph on the left summarizes the complete history of tritium activity in the pumped water.

Fig. 3A-5. Concentrations of tritium in the core samples from the L-100 borehole, May 1985.

We found no detectable increase in lithium or decrease in tritium levels. However, the data presented in Table 3A-1 show that bromide levels

- Rose significantly above the consistent background levels by 75 days after tracer injection,
- Peaked at a concentration approximately twice the background level and 750 times lower than the concentrations of the input tracer around 92 days after injection, and
- Returned to background by 168 days after the injection.

The time history of bromide concentration in deep-lysimeter samples taken in 1986 is shown in Fig. 3A-6. The travel time deduced from the bromide tracer, approximately 3 months, is within the range of times deduced from comparing tritium concentrations. The concentration of the tracer observed, less than 0.2% of the input concentration, explains why no reduction in tritium activity was seen as a result of the use of low-tritium water for the tracer; the variation in activity expected would be small compared with the 1% counting statistics for the tritium analyses.

The “unsaturated conditions” noted above, in combination with the shallow soil-moisture data, the tracer evidence for rapid vertical transport, and the results of other field and model studies by DRI, enhance our understanding of the recharge plume and its utility for possible future tracer work. For the purposes of tracer work and modeling, the quasi-two-dimensional plume is experimentally convenient. In addition, there are a wide range of unsaturated flow velocities within a short and easily measurable distance, and hydrologic transport occurs on a time scale well-suited to field experimentation.

Conclusions

The Cambric ditch and its associated recharge plume provide a unique and valuable opportunity to study the movement of water and radionuclides in alluvium, both as a result of gravity-driven flow and in the form of unsaturated, lateral migration in the vadose zone. Recharge transport has been directly measured at a rate of approximately 0.3 m/day. However, because of the distinctive and well-characterized tritium signature of RNM-2s water, we

are not limited by real-time observations. Migration times of up to 6 years have been determined by comparing tritium-activity trends in the vadose zone with the history of ditch activity.

Table 3A-1. Bromide concentrations in deep-lysimeter L-100 samples after injection of the surface tracer.

Date day	Julian injection	Days since (mg/L)	Br ⁻
3/26/86	85	0	0.15
3/31/86	90	5	0.15
4/10/86	100	15	0.15
4/21/86	111	26	0.17
4/28/86	118	33	0.15
5/15/86	135	50	0.17
6/09/86	160	75	0.27
6/17/86	168	83	0.32
6/26/86	177	92	0.34
7/07/86	188	103	0.32
7/22/86	203	118	0.27
8/04/86	216	131	0.23
9/03/86	246	161	0.20
9/10/86	253	168	0.16
10/09/86	282	197	0.14

Fig. 3A-6. "Breakthrough" of the bromide tracer released into the Cambric ditch, as shown in deep-lysimeter samples taken in 1986.

Section 3B. Vegetation Water Budgets

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Introduction

The unique environment created by the Cambic pumping experiment and described in Sec. 3A provides opportunities for using field tracers to study a variety of natural processes. Because of the long-term, constant irrigation of the desert floor, an extensively vegetated biological community has developed in and near the ditch and pond. The anomalous presence of a well-watered, heavily-vegetated desert community in the midst of an otherwise typically barren location leads directly to the supposition that the plants rely on the ditch water for most if not all of their water needs. Since this water contains a relatively high and time-dependent concentration of tritium, we had the opportunity to perform a variety of large-scale isotopic tracer studies of plant metabolisms, transpiration dynamics, and community water budgets in a natural setting. This section of the report describes the results of some initial hydrogen-isotope studies of vegetation water budgets.

The flux of water through living plants is of interest from a variety of standpoints. The uptake, retention, and release of water are important aspects of plant metabolism because they largely determine a plant's response to environmental stresses such as drought. Vegetational evapotranspiration can be an important factor in local and regional water budgets, with considerable impact on the hydrologic cycle. Finally, the fractionation and incorporation of hydrogen and oxygen isotopes in plant tissues is increasingly studied both as an aid to understanding plant metabolism and as a clue to past climate and environmental conditions that may have left their isotopic signatures in the durable portions of plant tissues.

Isotopic studies have considerable potential for elucidating plant metabolic processes. However, long-term or large-scale environmental studies are generally required to rely on the interpretation of small variations in naturally occurring isotope ratios (Epstein et al., 1977),

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whereas tracer studies are customarily limited to laboratory settings or to short time periods (IAEA, 1981).

To exploit the uniquely labeled environment of the Cambric ditch, from 1983 through 1985 LLNL and DRI cooperatively sampled and analyzed plants and other environmental media. To address a variety of specific questions, we

- Investigated the degree of isotopic equilibration between the water in the plant tissues and that in the ditch;
- Investigated the relationship between the isotopic ratios of natural deuterium (^2H) and anthropogenic tritium in the water present in the plants, ditch, and air;
- Studied the diurnal variations in the isotopic concentration of the water in plant tissues; and
- Compared the degree of tritium uptake in plants representing two different photosynthetic pathways (C_3 and C_4).

Methods

Initial reconnaissance samples were collected in May 1983, followed by an extensive systematic collection in August 1984. Samples taken in the initial collection were processed by two different methods, and replicate samples were collected and processed for methods comparison. Samples processed by LLNL were cut, bagged, and frozen on dry ice immediately after collection. They were kept frozen until the tissue water was extracted at LLNL by freeze drying. Samples collected by DRI were cut and immediately immersed in flasks of toluene; the flasks were then sealed and the contents subsequently distilled at DRI to remove the water fraction. Water samples were analyzed for tritium at LLNL by liquid scintillation spectrometry and for deuterium at DRI by mass spectrometry.

A second set of samples was collected in August 1984. Plants, which were sampled at different times throughout the day, included the cattail reed *Typha domingensis*, which was repetitively sampled at heights above the water of approximately 0.5, 1, and 2 m (1 to 2, 3 to 4,

and 6 to 7 ft, respectively). During this period, air-moisture samples were collected by drawing air through a tube of silica gel, which was subsequently baked out under vacuum in the laboratory to retrieve the water. Nighttime and daytime air-moisture samples were collected at two locations: at about 1 m above the ditch and within the vegetation canopy near where the reeds were being sampled, and approximately 92 ft south (upwind) of the ditch, also at a height of about 1 m.

Throughout the sampling periods, we regularly collected ditch-water and soil-moisture samples (the latter from lysimeters) and analyzed them for tritium.

Results and Discussion

Our results are presented in Appendix B. Included with these analytical data are species identifications and relevant location information. Other sections of the table contain, for the periods in question, data on the tritium content of the ditch water, soil moisture, and air moisture.

The results of methods-comparison samples (splits of the same plants processed by the two different methods) indicate that the two processes produce comparable results; the variability observed is believed to represent intra- and inter-specimen variability rather than systematic effects of the methods.

The transect samples (DRI samples 37 through 50, Appendix B, Tables B2 and B3) produced the expected results: samples in or immediately adjacent to the ditch showed elevated tritium levels, whereas at a distance of a few meters or more the plants showed little if any elevation above the local "background" level. However, the close-in samples did not exhibit tritium activities as high as the ditch-water samples, despite the fact that soil and lysimeter samples from similar locations led us to believe that the soil moisture should be very close in isotopic composition to the ditch water.

Figure 3B-1 summarizes additional results of the 1983 sampling effort. To check for systematic (diel) metabolic effects, we took samples in the early morning (low photosynthesis and

transpiration) and at midday (high photosynthesis and transpiration). Two species representing the C_3 photosynthetic pathway and two other species with a C_4 pathway were collected. In Fig. 3B-1, both deuterium and tritium data are presented relative to the ditch-water values for the same time; deuterium values are presented in terms of the Standard Mean Ocean Water (SMOW). Note that one vertical scale in each sub-figure is expressed as percent of ditch water, but that the scales are of very different magnitudes. Although there is no systematic difference in deuterium ratios, at both sampling times the C_3 plants show significantly lower tritium concentrations than the C_4 plants.

The water samples from plants are enriched in deuterium by a few percentage points relative to the ditch water, but are (in some cases) depleted in tritium by over 70%. The deuterium values are reasonable both in magnitude and in time pattern; plant respiration should result in modest enrichments of the liquid phase in the heavier isotope, and the magnitude of this effect should increase as the rate of transpiration increases during the day. Although the tritium values indicate that the plant contains "heavier" water at noon than in the early morning, the tritium is depleted rather than enriched relative to ditch water, and the differences are an order of magnitude greater than for deuterium.

To explore this effect more thoroughly, in 1984 we collected and analyzed additional samples. The plants selected were cattail reeds and salt cedar, both of which were rooted in the bottom of the ditch. Because they were growing out of the flowing water, there could be no question about their root exposure. In addition to further diel sampling, we also sampled the reeds as a function of their height above the surface of the water. Figure 3B-2 presents the results for the cattails and the salt cedar about 1 m above the surface of the water. Here again, we note substantial depletions relative to the tritium levels in the ditch and a trend toward increasing tritium levels (isotopically heavier water in the plants) over the course of the day.

Figure 3B-3 depicts the tritium activity in the cattail stems as a function of both time of day and height above the surface of the water. Air-moisture samples collected within the plant

Fig. 3B-1. Results of the 1983 sampling effort. Deuterium (a) and tritium (b) concentrations are shown for water extracted from plants having C_3 and C_4 photosynthetic pathways.

Fig. 3B-2. Tritium concentrations as a function of time of day in water extracted from cattail samples taken about 1 m above the surface of the water and from salt-cedar leaf samples.

Fig. 3B-3. Tritium activity in cattail water as a function of height above water and time of day. Vertical lines show the activities of the ditch water and of air moisture sampled at a location remote from the plant-sampling site.

growth a few feet above the water ranged in activity from about 150 pCi/mL at night to slightly less than 350 pCi/mL during the day.

If we assume that the same mechanisms affect both isotopes, the deuterium enrichments indicate that the reduction in plant-water tritium relative to the ditch water is not caused by any isotope-fractionation process. This leaves dilution as the sole reasonable candidate for consideration, and the only other reservoir of water available to the plants is the atmospheric moisture. This water source is similar in deuterium composition to the ditch and plant water but is initially free of tritium; exchange of water between the plant and the air could therefore dilute the plant-water tritium without dramatic effect on the deuterium composition.

The data permit some rough estimates of the relative amounts of water from the two sources in the plant tissues. If we ignore the effects of isotopic fractionation and treat the remote air and the ditch water as the two end members, the results indicate that approximately 50% of the nighttime water inventory of the cattails and a similar but slightly smaller fraction of the salt-cedar water are derived from the atmosphere. This is a lower limit on the amount of atmosphere-derived moisture, because the tritium activity of the air moisture in the immediate vicinity of the plants is higher than the remote values as a result of local evapotranspiration of the ditch water. The effect of more rapid plant transpiration during the daylight hours is to raise the fraction of ditch water in the total plant tissue to 90% or more, presumably by increasing the rate of transpiration outflow relative to the exchange and back-diffusion of atmospheric moisture at the air-plant interface. The plant data of Fig. 3B-1 show that air moisture must account for up to 70% of the water content of some of the plants growing immediately adjacent to the ditch.

Conclusion

The results are consistent with laboratory studies of HTO-vapor uptake by plants, which indicated that plant water reached tritium values that were 20 to 50% of the vapor-phase values (IAEA, 1981, p. 44). However, the results are somewhat surprising in this case, in view of the low ambient humidity and the ready availability of labeled water from the saturated or nearly

saturated soil system. The data do not permit us to identify the mechanism in detail, but they suggest that the air-to-plant exchange of moisture must have rates that can be a significant fraction of the plant's total net water-transpiration flux.

The labeled ecosystem associated with this experiment offers the opportunity for more detailed study of water fluxes and exchange rates, and the results further suggest that interpretation of plant water and tissue must include consideration of possible differences among the isotope ratios in water in the air, soil, and ground.

Section 4. Technetium Migration in Nevada Test Site Groundwater

Contributors: R. J. Silva, R. Evans, J. H. Rego, and R. W. Buddemeier*

Introduction

^{99}Tc is produced in the fission process and is therefore present in both spent nuclear-reactor fuel and in debris from nuclear tests. It is a low-energy beta emitter with a maximum energy of 292 keV and a half-life of $2.13 \pm 0.05 \times 10^5$ y (Kocher, 1981). Because of its long half-life, high solubility, and low sorptive properties under oxidizing conditions, ^{99}Tc is considered a potential hazard if released to environmental waters (Luxenburger and Schuttelkopf, 1984).

Isherwood (1985) and Buddemeier and Isherwood (1985) used the EQ3/6 geochemical computer code developed by LLNL's Earth Sciences Department to predict the chemical speciation and solubility of technetium in groundwater. The groundwater at NTS tends to be oxidizing ($E_h > 250$ mV) and to have pH values between 7 and 9 (Buddemeier and Isherwood, 1985; Ogard and Kerrisk, 1984). It was predicted that TcO_4^- would be the major solution species in these waters, and that technetium concentrations would not be limited by solubility. Because nuclear weapons are tested underground, technetium should be present in small concentrations in some of the water sampled by the Radionuclide Migration (RNM) Project (Isherwood, 1985; Buddemeier and Isherwood, 1985). Laboratory measurements conducted under oxidizing conditions indicate that technetium is not sorbed to any extent in crushed-rock samples from NTS (Daniels and Thompson, 1984). Thus, one would expect technetium to be rather mobile in the environment and to follow tritium rather closely in its migration behavior.

Isolation Technique

For two reasons, we needed to develop a new way to chemically isolate and concentrate the ^{99}Tc before we could perform reliable analyses. First, because there are no gamma rays emitted in the decay of ^{99}Tc , the low-level gamma spectroscopy methods we usually use to

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analyze NTS samples could not be applied. Second, beta-counting techniques, although applicable, are not as specific as the gamma-ray methods, making chemical analysis necessary for positive isotopic identification.

Therefore, in 1985, we developed and tested a fast and simple chemical-separation scheme for isolating ^{99}Tc from liter quantities of RNM samples. The method is based on a more complex scheme reported by Luxenburger and Schuttelkopf (1984) in which the concentration of ^{99}Tc is determined by liquid-scintillation counting. The steps for testing a 1-L sample are as follows:

- A 1-L sample is acidified and made oxidizing by the addition of 10 cm³ of concentrated HCl and 5 cm³ of 30% H₂O₂. (The HCl and H₂O₂ are scaled up for larger sample volumes.) The solution is stirred with a Teflon-coated magnetic bar for about 1 h.
- About 0.5 g of AG 1 × 8 (100- to 200-mesh) anion resin is added to the solution. (The same amount of resin is added for all sample volumes.) The stirring is continued for 4 h or longer, and the resin is then allowed to settle for 1 h or longer.
- The clear supernate is decanted and the resin is flushed into a 0.5-cm-diam by 10-cm-long glass column. The resulting resin bed is about 4-cm high.
- The resin bed is washed first with 2 cm³ of 0.1M HCl and then with 2 cm³ of distilled, deionized water. (If large amounts of tritium are present, additional water washing is necessary.)
- The resin is converted from the chloride form to the perchlorate form by passing 2 cm³ of 1M NaClO₄ through the resin.
- The technetium is reduced and eluted from the column by passing a 1M NaClO₄-0.02M Na₂SO₃ solution (pH = 12) through the resin.
- Two-cm³ fractions of eluent are collected in glass counting vials, 15 cm³ of scintillation cocktail is added, and the mixture is counted in a liquid-scintillation counter.

Preliminary Investigation

The first preliminary tests of this method were reported in 1986 (Silva et al., 1986). In summary, samples were tested from the sites of the Cheshire, Bilby, Nash, and Cambric Events. Although ^{99}Tc was found in most of the samples, none exceeded the maximum permitted concentration for ^{99}Tc of $3 \times 10^{-4} \mu\text{Ci/mL}$, as given in the United States Nuclear Regulatory Commission (USNRC) *Code of Federal Regulations* (1982). The results from ultrafiltered and normally filtered samples from the Cheshire site showed that the technetium did not appear to be associated to any appreciable extent with particulate material and could be considered primarily dissolved. These results suggested that technetium was indeed present as TcO_4^- , as predicted. However, some of the preliminary results for ^{99}Tc concentrations near the detection limit were inconsistent for the Nash and Cambric samples. Tests performed a few months later on similar but larger-volume samples (thus at higher sensitivities) from these two sites showed no indication of technetium.

Recent Investigation

In early 1987, an improved analysis scheme with $^{95\text{m}}\text{Tc}$ as a chemical yield tracer was included in the processing of a second series of samples in order to isolate and determine the concentration of ^{99}Tc in NTS groundwaters (Silva et al., 1988). A secondary goal was to confirm that technetium was not associated with filterable material. Samples from the Faultless site were included in this series.

Test Procedure

Archived samples (in 1- to 4-L volumes) of selected groundwaters from the Cheshire, Bilby, Nash, Bourbon, Cambric, and Faultless Event sites were used in the study. All reagents were of analytical grade and solutions were made with distilled, deionized water. The ^{99}Tc standard used to calibrate the liquid scintillation counter and to test the separation procedures was obtained from and certified by the U.S. National Bureau of Standards. Figure 4-1(a) shows a

beta-energy spectrum obtained with the liquid-scintillation counter from an aliquot of the stock solution. The ^{99}Tc standard was counted with a $90.7 \pm 1.2\%$ efficiency.

We followed the same chemical separation procedure used for our preliminary migration study except that now the solutions were "spiked" with a known quantity of $^{95\text{m}}\text{Tc}$ tracer. The $^{95\text{m}}\text{Tc}$ ($T_{1/2} = 61$ d) was produced by using the Lawrence Berkeley Laboratory's 88-in. cyclotron to irradiate a 0.003-in.-thick niobium foil with 30-MeV alpha particles (Bond and Jha, 1970). After dissolution of the foil in 5 cm³ of a mixture of 27M HF and 1M HNO₃, the solution was diluted to 100 cm³, a stoichiometric amount of H₃BO₃ was added to complex the fluoride ions, and the solution was processed in the same manner described earlier for the NTS samples. Figure 4-1(b) shows an energy spectrum obtained with the liquid scintillation counter from an aliquot of the $^{95\text{m}}\text{Tc}$ stock solution.

Data Analysis Procedure

The NaClO₄ and the first three NaClO₄-Na₂SO₃ 2-cm³ fractions eluted from the anion column for each sample were counted for 1000 to 2000 minutes in a liquid scintillation counter. The resulting 1000-channel energy spectra were collected, and the background was subtracted. A typical spectrum for the Cheshire sample and a typical background spectrum are shown in Figs. 4-2(a) and 4-2(b), respectively. The concentrations of ^{99}Tc and the yield tracer $^{95\text{m}}\text{Tc}$ were determined by spectra-stripping techniques using standard spectra for these isotopes. The ^{99}Tc and $^{95\text{m}}\text{Tc}$ were finally identified from their characteristic energy spectra and their column elution behaviors; i.e., both energy spectra (after background subtraction) and elution behavior were required to be the same as those determined for the standards of these isotopes. The detection limit is 9×10^{-7} μCi for the 1-L samples, or 2.3×10^{-7} μCi for the 4-L samples.

Results

Using ^{99}Tc standards, we found that on the average $93.0 \pm 7.0\%$ of the tracer was recovered and that $87.0 \pm 5.0\%$ of the tracer was eluted in the first 4 cm³ of eluent. We thus had confidence in the relative accuracy of our results. Figure 4-3 shows a typical elution curve.

Fig. 4-1. Beta-energy spectra obtained with a liquid scintillation counter (a) for the ^{99}Tc stock solution, and (b) for the $^{95\text{m}}\text{Tc}$ stock solution.

Fig. 4-2. Typical energy spectra (a) for the Cheshire sample, and (b) for the background.

Fig. 4-3. Typical elution behavior of ^{99}Tc from an anion resin column (AG 1 $\times$ 8, 100-200 mesh, 0.5-cm diam by 6-cm long).

The results of the analyses of samples obtained in and near the six event sites are shown in Table 4-1. The error limits represent one standard deviation and result from uncertainties associated with chemical yields, counting, and data analysis. The tritium contents, corrected back to the time of the event ($T = 0$), are included for comparison.

Conclusions

^{99}Tc was detected in water samples taken in or near the cavity regions of the Cheshire, Bilby, and Faultless Event sites. However, none exceeded the maximum permissible concentration for water of $3 \times 10^{-4} \mu\text{Ci/mL}$ as given in the Code of Federal Regulations (USNRC, 1982). None of the satellite well waters contained ^{99}Tc above our detection limit. The ^{99}Tc detected in the Faultless sample is the first radionuclide other than tritium reported in water from this event site (see Buddemeier and Isherwood, 1985, for a description of the Faultless site and previous sampling efforts there).

Laboratory measurements conducted under oxidizing conditions yield K_d values near zero for technetium (Daniels and Thompson, 1984; Serne and Relyea, 1982). Thus, one would expect technetium to follow tritium rather closely in its migration behavior. Because water samples can be obtained from both the cavity area (RNM-1) and a satellite well (RNM-2), the Cambria site would seem to provide a unique opportunity for testing migration patterns in the field. Unfortunately for this purpose, none of the Cambria samples indicated ^{99}Tc above our detection limit. Samples obtained from the cavity did have higher-than-normal detection limits. These were caused by the large amount of tritium in these waters which, because it could not be totally removed during the chemical processing, caused background subtraction problems.

The results for ^{99}Tc , obtained from Cheshire samples passed through filters of different pore sizes, confirm our earlier expectations that technetium would exhibit a behavior similar to Na and Sb. There was not clear evidence that technetium is sorbed to any appreciable extent on filterable material in the water samples. This result could be expected if the technetium is

indeed present as TcO_4^- as predicted. Most naturally occurring colloids, e.g., clays, iron hydroxides, etc., have a negative surface charge at neutral and alkaline pH values (Yariv and Cross, 1979). These colloidal particles would tend to sorb positively charged solution species and not negatively charged ones (Yariv and Cross, 1979).

Table 4-1. Results of technetium analyses.							
Experimental site	Sample location	Sampling date	Sample volume (L)	Filter size (μm)	^{99}Tc		^3H ($\mu\text{Ci/mL}$ at T_0)
					Count ($\mu\text{Ci/mL}$)	% SD	
Cheshire (U20n)	Cavity	5/09/85	1.17	0.003	5.11×10^{-8}	11.5	5.0×10^{-1}
		10/23/84	1.00	None	4.90×10^{-8}	10.6	4.1×10^{-1}
	Above cavity	5/29/85	1.00	0.05	3.83×10^{-8}	12.5	4.3×10^{-1}
		5/29/85	1.20	0.003	2.73×10^{-8}	11.6	4.3×10^{-1}
Bilby (U3cn)	Chimney	7/19/82	1.00	0.45	4.05×10^{-8}	5.0	9.9×10^{-2}
		6/18/85	1.00	0.20	5.87×10^{-8}	4.0	9.5×10^{-2}
Faultless (UCP1P2SR)	Well (2590 ft) (2390 ft)	7/21/83	3.10	0.45	4.77×10^{-9}	10.4	5.0×10^{-2}
			1.75	0.45	1.49×10^{-9}	12.1	5.0×10^{-2}
Nash (U2ce)	Satellite well	4/18/84	4.0	0.45	$< 2.3 \times 10^{-10}$		6.3×10^{-2}
Bourbon (U7n)	Satellite well	6/24/83	4.0	0.45	$< 2.3 \times 10^{-10}$		$< 3.3 \times 10^{-6}$
Cambric (U5e)	Lower cavity	1/06/75	1.0	0.45	$< 9.0 \times 10^{-10}$		9.9
	Chimney	8/07/75	4.0	0.45	$< 2.3 \times 10^{-10}$		1.4×10^{-1}
	Chimney & adjacent	4/10/85	4.0	0.20	$< 2.3 \times 10^{-10}$		7.2×10^{-5}
	Satellite well	11/8/82- 11/10/82	3.5	0.45	$< 2.3 \times 10^{-10}$		6.3×10^{-3}

Section 5. Other Studies

Section 5A. Further Data on 1974 Study of Radionuclide Migration at Yucca Flat

Contributors: J. H. Rego, R. W. Buddemeier, and K. V. Marsh

Introduction

Early in 1986, LANL reported unexpected tritium and fission product activity in the water flowing into the proposed emplacement hole for the Aleman test (Area 3, Yucca Flat). In order to relate these results (Sewell, 1987) to earlier observations of apparent rapid transport of radionuclides, we reviewed the earlier work done at LLNL and published by Crow (1976).

Crow (1976) reported on the observations of tritium in three holes in Area 2 (see Frontispiece). The first hole, U2aw, was found to have an initial inflow of 28.6 m^3 of groundwater per day, with tritium concentrations ranging from 2 to $12 \times 10^{-3} \text{ } \mu\text{Ci/mL}$ over the period of observation (April through September, 1974). Water from this hole had also been analyzed for gamma emitters with negative results, but the analytical procedure and sensitivity were not reported. The suspected source of the tritium was the Commodore Event, which had been fired on 20 May 1967 in hole U2am, 465 m south east of U2aw. Water from the second exploratory drill hole (UE2aw) also had an elevated tritium concentration (approx. $10^{-4} \text{ } \mu\text{Ci/mL}$). A third exploratory hole, UE2ar, was drilled 981 m south of U2aw and 361 m from a second possible source, the Agile Event, that had been fired in February 1967 with a working point at 733 m.

Crow (1976) reported that four water samples were bailed from hole UE2ar on May 10, 1975. At the time of sampling, the hole had caved in nearly to the level of the water table (544 m below ground surface). The samples showed tritium activities ranging from 1.1 to $5.0 \times 10^{-4} \text{ } \mu\text{Ci/mL}$, with an average of $3 \times 10^{-4} \text{ } \mu\text{Ci/mL}$. Crow concluded that Agile was the probable source of the tritium observed in UE2ar.

Experimental

A search of the Nuclear Chemistry Division's sample archives located four 1-L sample bottles labeled " H_2O -UE2ar-1790 ft-5/10/75," "(1 of 4)" through "(4 of 4)." The sample liquid

was brown and appeared to contain considerable sediment. Sample (3 of 4) was gamma counted without filtration. Sample (4 of 4) was centrifuged, filtered through a 0.45- μ m filter and gamma counted as (a) filter+sediment and (b) filtrate. The filtrate was also analyzed for tritium. The filtrate and the unfiltered, sediment+water samples were counted in a Marinelli beaker (see Sec. 5B); the filter+sediment sample was counted in a standard 25 cm³ counting vial.

Results and Discussion

Table 5A-1 lists the radionuclide concentrations observed; all have been decay corrected to the 1975 sampling date. The tritium value is within the range previously measured (Crow, 1976). It is significant, however, that ¹²⁵Sb was found in the filter+sediment sample and ¹³⁷Cs was found in all three sample fractions analyzed. These nuclides were not originally detected and were not found in the U2aw groundwater; they are, however, observed to undergo hydrologic transport at the Cheshire site and in Area 3.

It is interesting to note that for both of the gamma-emitting nuclides detected in the UE2ar samples, the largest fraction of the total activity was found in the filterable materials rather than in the filtrate. The samples from Cheshire (see Sec. 2) indicate that antimony is almost completely dissolved and cesium is predominantly so. However, the UE2ar samples are substantially older than the Cheshire samples, both in terms of the time elapsed since detonation and in terms of the time stored as a mixed sediment+water system. These results hint at the possibility that the kinetics of sorption equilibrium for these species are very slow, producing different distribution coefficients as a function of time over periods much longer than those of typical lab or field experiments. Another possibility, of course, is that the chemical characteristics of either the groundwater or the solids in Area 2 are very different from conditions near the Cheshire site; we know, for example, that groundwater temperatures on Pahute Mesa are significantly higher than in Yucca Flat. As we concern ourselves with longer-term migration issues, these matters would seem to merit further attention.

Table 5A-1. Radionuclides in groundwater samples from hole UE2ar at NTS: comparison between measurements made in 1975 and recent measurements corrected to 1975.

Nuclide	Recent measurements			Range of 1975 measurements ^a (10 ⁻⁴ μCi/mL)
	Sample (3 of 4)	Sample (4 of 4)		
	Nonfiltered (μCi/mL/% error)	Filter + sediment (μCi/mL/% error)	Filtrate (μCi/mL/% error)	
³ H	NA ^b	NA	2.68 × 10 ⁻⁴ /1.0	1 to 5
⁴⁰ K	2.81 × 10 ⁻⁷ /6.17	1.40 × 10 ⁻⁷ /5.6	1.6 × 10 ⁻⁷ /50	NA
¹²⁵ Sb	ND ^b	6.02 × 10 ⁻⁸ /16.4	ND	NA
¹³⁷ Cs	2.14 × 10 ⁻⁸ /5.55	5.67 × 10 ⁻⁸ /1.3	1.29 × 10 ⁻⁹ /51.4	NA

^a From Crow (1976).

^b NA = not analyzed; ND = not detected.

If the source of the UE2ar activity is the Agile Event, and if the transport mechanism is hydrologic migration, the indication is that some radionuclides are being transported in tuff more rapidly than at first expected. A transit time of 8 y or less over a distance of about 360 m implies a migration rate of at least 45 m/y. Initial estimates of groundwater flow velocities were in the range of 30 m/y (Crow, 1976).

Conclusions

The results of our review of earlier work and our examination of preserved groundwater samples tend to reinforce the conclusions drawn from the Cheshire site and Area 3 observations, i.e., that antimony and cesium are relatively mobile and may be expected to undergo reasonably prompt migration with moving groundwater. The Area 2 and Area 3 results, obtained from regions of tuffaceous alluvium, raise the question of why similar radionuclide movements have not been seen in the induced-migration experiment at the Cambric site.

Section 5B. Calibration and Applications of Marinelli Beakers

Contributors: R. A. Failor, J. Beiriger, J. H. Rego, R. W. Buddemeier, and K. V. Marsh

Introduction

Marinelli beakers are frequently used to obtain preliminary analyses of very low-level, large-volume samples, especially liquids. The GAMANAL program (Gunnink and Niday, 1972) is not able to handle the geometry of these beakers, so special efficiency calibrations are

necessary. We have performed detector-specific calibrations for our systems that allow us to account for counting geometry and for gamma-ray absorption effects on both the sample and background activities.

Initially, we intended to use the Marinelli counting method to scan incoming samples to detect gross differences in concentration and to choose samples for more precise workup. However, we found that a simple calibration allowed the Marinelli beakers to be used for semi-quantitative analyses. In this discussion, we include results from an analysis of samples from the Cheshire site.

Calibrations

Procedure. We used a National Bureau of Standards (NBS) standard reference solution containing ^{125}Sb , $^{125\text{m}}\text{Te}$, ^{154}Eu , and ^{155}Eu as the calibration solution. NBS suggests that this solution be used as the mixed-radionuclide solution standard for efficiency calibration of germanium-spectrometer systems.

The reference solution was diluted to produce a reduced-activity solution in sufficient volume to fill two 1-L Marinelli beakers (manufactured by GA-MA Associates, Inc.) and to provide 50 mL for small-volume measurements in a standard geometry.

The activity of the diluted solution was measured using the small-volume samples and compared to the values calculated from the NBS reference values. The measured and calculated values agreed to within less than 3%. Duplicate counts were made of both Marinelli beakers containing the diluted standard. These values agreed to within counting uncertainty (<5%).

To account for possible absorption of ambient gamma rays by the water-filled Marinelli beakers, long background measurements were made for each detector with and without the Marinelli beaker in place.

Results. The energy-dependent efficiency was calculated for each detector separately. Reciprocal efficiency as a function of energy is shown in Table 5B-1 and plotted in Fig. 5B-1. The deviations from linearity at low energies can be accounted for by absorption of the gamma

Table 5B-1. Details of gamma-ray Marinelli calibration.				
Radionuclide	Photon energy (keV)	(Efficiency) ⁻¹		
		Det L-01	Det E-01	Det F-01
¹⁵⁵ Eu	86.6	58	59.64	47.49
¹⁵⁵ Eu	105.3	41.09	50.17	42.68
¹⁵⁴ Eu	123.1	53.73	49.2	43.85
¹²⁵ Sb	176.4	47.16	50.4	45.89
¹⁵⁴ Eu	280	64.59	72.69	68.15
¹²⁵ Sb	380.5	81.8	100.4	91.4
¹²⁵ Sb	427.9	91.42	112.4	105
¹²⁵ Sb	463.9	98.24	122.2	113
¹⁵⁴ Eu	591.7	135.6	169.6	160.2
¹²⁵ Sb	600.6	122.6	154.6	145.3
¹²⁵ Sb	635.9	129.4	164.6	154.6
¹⁵⁴ Eu	723.3	156.8	198.2	186.3
¹⁵⁴ Eu	873.2	184.8	237.7	224.7
¹⁵⁴ Eu	996.4	196.6	254.1	235.5
¹⁵⁴ Eu	1004.8	196.2	258.7	243.3
¹⁵⁴ Eu	1274.4	235.9	317.1	299.2
¹⁵⁴ Eu	1596.5	250.8	350.5	327.7

^a (Efficiency)⁻¹ = photons/min emitted ÷ counts/min observed.

Fig. 5B-1. Marinelli counting efficiency for three detectors.

rays in the sample. The deviations from linearity at the high energies are due to summing of the gamma rays from a given isotope. Summing corrections for the NBS nuclides resulted in linear high-energy values. Although this method gives us a general calibration of efficiency versus gamma-ray energy, it is not applicable to radionuclides from which cascade gamma rays produce large summing effects. We intend to perform specific empirical calibrations for all such nuclides of interest to the HRMP program.

The absorption of the gamma rays by a Marinelli beaker filled with water also results in a reduction in the background activity seen by the detector. We have measured the effect for each detector. At energies below 1 MeV, the changes were up to 10%. This was greater than the counting uncertainty. For this reason, we used "Marinelli background values" for all the analyses presented here.

Analysis of Cheshire Samples

To investigate the association of radionuclides with colloidal particles, as well as the size distribution of small particles (<1.0 to $>0.003\ \mu\text{m}$), we ultrafiltered two 1-L samples and used the calibrated Marinelli beakers to analyze the filtrates. Results for these two samples (identified as $0.003\text{-}\mu\text{m}$ filter size) were compared with those for nonfiltered and conventionally filtered samples and with the results of large-volume sample processing.

Table 5B-2 shows the results of this experiment. If we compare the ^{125}Sb concentrations for nonfiltered and ultrafiltered 1-L portions of sample B-8, we see no significant difference in the values. This information supports the results obtained from large-volume samples (see Sec. 2 and Appendix A); namely, ^{125}Sb in these samples is almost completely dissolved ($>97\%$). The same comparison for ^{137}Cs shows a difference of 60%, indicating that 60% of the activity is in solution (passing through a $0.003\text{-}\mu\text{m}$ membrane) and 40% of the activity is attached to particles greater than $0.003\ \mu\text{m}$. This is consistent with the $0.003\text{-}\mu\text{m}$ cutoff observed from evaporation of our large-volume (194.5-L) B-8 sample, which excluded about 35% of the total ^{137}Cs .

The remainder of the table presents the results of Marinelli beaker counts of other samples passed through filters of various pore sizes. They are included to indicate the general accuracy

Table 5B-2. Results of calibrated Marinelli beaker scans of water samples from the Cheshire site ($\mu\text{Ci/mL}$).

Date collected	09/—/76	09/09/83	09/09/83	10/23/84	10/23/84	05/09/85	05/09/85	05/25/85	06/19/85	09/11/85	10/01/85
Detector	L-01	L-01	L-01	L-01	L-01	F-01	E-01	L-01	L-01	L-01	L-01
Sample label	S-0	S-1	S-1	B-3	B-4	B-8	B-8	S-38	S-31	S-36	S-37
Sample size (L)	1.0169	0.950	0.950	0.980	0.898	1.0366	1.03412	0.998	1.0124	0.9231	1.063
Filter cutoff (μm)	(None)	0.45	0.006	0.20	0.003	(None)	0.003	(None)	(None)	0.45	0.45
Nuclide	Cavity						Formation				
^{22}Na	ND ^a	1.11×10^{-8} 55% ^b	1.13×10^{-8} 26%	3.31×10^{-8} 17.8%	1.47×10^{-8} 33.2%	6.09×10^{-8} 20.3%	ND ^a	ND	ND	ND	ND
^{40}K	5.06×10^{-5} 8.0%	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
^{85}Kr	ND	8.17×10^{-5}	ND	5.22×10^{-6} 28.1%	ND	ND	ND	2.62×10^{-5} 6.6%	2.40×10^{-5} 7.2%	1.83×10^{-5} 1.5%	2.54×10^{-5} 1.3%
^{125}Sb	1.78×10^{-6} 1.7%	8.81×10^{-6} 4%	7.74×10^{-6} 4%	5.74×10^{-6} 1.4%	6.17×10^{-6} 1.2%	5.14×10^{-6} 1.8%	5.17×10^{-6} 1.8%	3.75×10^{-6} 1.2%	3.97×10^{-6} 1.0%	2.67×10^{-6} 1.3%	3.46×10^{-6} 1.2%
^{137}Cs	5.13×10^{-9} 9.2%	2.06×10^{-7} 2%	1.62×10^{-7} 2%	3.94×10^{-6} 1.6%	2.87×10^{-6} 1.3%	3.42×10^{-6} 0.4%	2.03×10^{-6} 0.5%	4.30×10^{-7} 1.2%	7.32×10^{-7} 1.2%	2.25×10^{-7} 1.3%	6.41×10^{-7} 1.2%

^a ND = not detected; no limit calculated.

^b % uncertainties are based on counting statistics, ± 1 standard deviation.

and limits of detection of the technique. The ^{85}Kr results merely indicate the concentration present in the sample when counted; no attempt was made to take or preserve these samples for quantitative determination of the ^{85}Kr .

Conclusion

This technique has been shown to be useful and accurate in its present form, but can be refined considerably (Zimmer, 1980), and work is in progress to do so. At present, we must take the output of a computer analysis of the spectrum calculated as counts per minute (cpm) of each gamma ray and, using plots similar to that shown in Fig. 5B-1, perform calculations by hand to convert the cpm to the desired concentration units. In the future, we will incorporate in the computer program a table of "pseudo-branching intensities" that will permit the GAMANAL code to identify isotopes and calculate concentrations directly. These pseudo-branching intensities will combine the actual nuclear decay branching intensities with correction factors for absorption of the gamma rays by the sample and for coincidence summing effects. In the meantime, the disposable Marinelli beakers give us a very fast and convenient means of analyzing up to 1-L volumes of liquid with accuracy and sensitivity suitable for many HRMP applications.

Section 5C. Determination of Particulate Activity: Bulk Filters versus Ashed Filters

Contributors: J. H. Rego, R. W. Buddemeier, and K. V. Marsh

Introduction

Standard LLNL procedures for the determination of particulate activity in large-volume water samples rely on direct counting of the filters used to process the sample. The filters representing each size fraction are drained, cut or folded as necessary, and then packed into a standard container (usually a sealed aluminum can, 8.5 cm i.d. by 4 cm i.h., commonly referred to as a "tuna can"). These containers are counted directly on low-background Ge(Li) detectors, and the gamma-ray activity is calculated using the GAMANAL code (Gunnink and Niday, 1972). The process is convenient, involves little risk of sample loss or contamination, and

provides data on the distribution of activity as a function of particle size. However, there are at least two potential disadvantages, both related to the extended-source geometry of the tuna can. First, because an extended-source geometry reduces counting efficiency relative to smaller sources, sensitivity is reduced. Second, we know that the filter material is not uniformly distributed throughout the tuna can, and we have every reason to believe that the particulate material is not uniformly distributed on the filter. The extended source is therefore inhomogeneous to some degree, but the GAMANAL code necessarily treats it as a homogeneous source.

For studies focused on the distribution of activity between the dissolved and total-particulate phases, it seemed clear that combined filters reduced to a relatively small volume would provide more favorable geometry (in terms of both size and homogeneity) and higher total activity per sample. In principle, this should result in greater sensitivity and more accurate determinations of total particulate activity. To test these hypotheses, we compared the activities obtained the traditional way, by counting filters directly, with the activities measured by counting ashed composites obtained by ashing and then combining all of the ashed filters from a given sample.

Procedures

The samples used for the comparison were the filters from Cheshire samples B-1 through B-6 (see Sec. 1 and Appendix A), all of which had received long counts on low-background detectors in the standard tuna-can configuration.

The sample cans were opened, and the filter pieces were placed in a porcelain crucible and heated in a small muffle furnace at about 475°C for 8 hours or until the filters were completely ashed. The ashed filter fractions were then combined into a composite for each sample, and the composite ash sample was counted on a low-background Ge(Li) gamma-ray spectrometry system. A set of unused filters was also ashed and counted to determine whether the filters themselves contributed to the measured activities.

For purposes of comparison, the activities of the individually counted unashed filters were summed for each sample, and the percent standard deviation of the sum was calculated according to the formula

$$\%SD = \frac{[(SD_1)^2 + (SD_2)^2 + (SD_3)^2 \dots]^{1/2}}{(A_1 + A_2 + A_3 + \dots)} \times 100,$$

where %SD = the percent standard deviation of a given activity A, and SD = (%SD × A)/100. Limit values were omitted from the sums when discrete values were available for any of the fractions, but when only limits were available, they were summed and their sum was listed as an upper limit for the total activity.

Results

Complete data for the individual filters and ashed composites of samples B-1 through B-6 are tabulated in Appendix A. Table 5C-1 presents a summary of the summed-filter and ashed-composite results for the major radionuclides in all six samples and in a set of unused filters. The sample counts were corrected to activity at time zero (T_0) for the detonation; the counting data for the unused filters were presented as activity (limits) at the time of the count, since nearly all the activity reflects background counts and therefore would not vary with time. Contributions to the sample activities by the unused filters can be seen to be negligible.

Table 5C-2 presents for each of the major nuclides in each sample the ratio of the ashed-composite activity to the summed-filter activity. The uncertainties associated with these ratios are calculated from the counting uncertainties in Table 5C-1. To facilitate comparisons, two further sets of calculations are presented in Table 5C-2: at the bottom of the table are the means of the ratios averaged across all nuclides in a give sample, and in the right-hand column are the means of the ratios averaged across all samples for a given nuclide. In these two cases, the standard deviations presented are calculated from the set of ratios averaged and do not reflect the counting statistics. For reasons discussed below, ratios with a compounded

counting uncertainty in excess of 10% were excluded for the calculation of the means; these values are marked with a superscript "c" in the table.

Several conclusions can be based on Table 5C-1. First, we note that the summed and ashed values are generally reasonably close. We can also see that the ashed sample values usually have lower counting uncertainties, and that the difference in the counting uncertainties increases as the level of activity decreases (see, for example, the europium isotopes and ^{60}Co in samples B-5 and B-6). Related to this is the observation that sensitivities are improved by counting the ash; several nuclides that are shown only as limits in the sums appear as real (albeit very uncertain) numbers in the ash counts.

The ashed-composite samples from both B-5 and B-6 were counted twice in order to obtain an estimate of the reproducibility of the results for an individual ash sample; the data are presented in Appendix A and in Table 5C-3. For the major nuclide activities and ratios listed in Table 5C-3, the mean absolute difference between counts was $3 \pm 9\%$ for the B-5 comparison and $11 \pm 10\%$ for the B-6 comparison.

Table 5C-2 indicates that the ashing procedures introduced no major biases into the relative concentrations of the nuclides in a given sample. Nuclide yields from the ashed samples (relative to the summed values) were reasonably consistent within a sample; results for B-1, B-2, and B-5 indicate good total recovery; results for B-4 and possibly B-3 suggest the possibility of sample loss; and the few reasonably precise values for sample B-6 indicate that the values for the ashed sample were higher.

Discussion

A review of the process suggests that, if there are any predictable differences between the values for the summed and ashed-composite samples, one would expect the ash values to be lower. The two possible sources of loss are in the increased manipulation involved in transferring and combining the relatively small ash samples and in the possibility of loss by spattering or volatilization. There are no systematic differences between the refractory nuclides (e.g., the lanthanides) and those which might be imagined to have some potential for volatilization

Table 5C-1. Activity comparisons of directly counted and ashed filters ($\mu\text{Ci}/\text{mL}$ at T_0).^a

Sample ID Collection date Volume (l)	Cheshire B-1 9/8/83 188				Cheshire B-2 9/9/83 200				Cheshire B-3 10/23/84 166				Cheshire B-4 10/23/84 194.5			
	Summed fractions	% SD ^b	Combined ashed	% SD	Summed fractions	% SD	Combined ashed	% SD	Summed fractions	% SD	Combined ashed	% SD	Summed fractions	% SD	Combined ashed	% SD
⁵⁴ Mn	7.4×10^{-8}	12.0	5.8×10^{-8}	34	4.1×10^{-8}	23.9	2.9×10^{-8}	46.4	NL ^c	—	1.2×10^{-8}	64	3.3×10^{-8}	45	3.3×10^{-8}	67
⁶⁰ Co	4.0×10^{-9}	1.1	3.6×10^{-9}	0.7	2.0×10^{-9}	2.6	1.9×10^{-9}	0.9	4.6×10^{-10}	6.4	3.7×10^{-10}	1.5	2.4×10^{-9}	3.6	1.9×10^{-9}	0.9
¹⁰⁶ Ru	4.3×10^{-6}	2.3	3.7×10^{-6}	2.5	2.7×10^{-6}	3.0	2.5×10^{-6}	4.8	9.5×10^{-7}	8.2	7.4×10^{-7}	3.4	7.1×10^{-6}	4.4	5.7×10^{-6}	2.9
¹²⁵ Sb	6.6×10^{-8}	2.2	6.1×10^{-8}	1.5	4.4×10^{-8}	3.1	4.5×10^{-8}	1.7	2.6×10^{-8}	2.2	2.1×10^{-8}	1.6	1.3×10^{-7}	2.2	1.1×10^{-7}	1.2
¹³⁷ Cs	3.9×10^{-7}	0.8	3.7×10^{-7}	0.8	7.2×10^{-7}	0.9	6.7×10^{-7}	1.1	1.0×10^{-7}	0.9	9.5×10^{-8}	0.8	1.2×10^{-8}	1.0	1.0×10^{-8}	0.7
¹⁴⁴ Ce	4.9×10^{-9}	7.8	5.2×10^{-9}	7.1	3.1×10^{-9}	10.7	3.8×10^{-9}	25.6	5.1×10^{-10}	17.5	8.5×10^{-10}	12.5	6.7×10^{-9}	15.9	5.6×10^{-9}	8.5
¹⁵² Eu	3.8×10^{-9}	1.9	3.5×10^{-9}	1.2	2.4×10^{-9}	3.7	2.4×10^{-9}	1.3	7.4×10^{-10}	5.9	6.0×10^{-10}	2.4	5.3×10^{-9}	2.5	4.2×10^{-9}	1.2
¹⁵⁴ Eu	6.7×10^{-9}	1.4	6.2×10^{-9}	1.2	3.2×10^{-9}	3.2	4.2×10^{-9}	1.1	1.2×10^{-9}	4.0	1.1×10^{-9}	1.8	9.1×10^{-9}	2.1	7.2×10^{-9}	1.0
¹⁵⁵ Eu	1.9×10^{-8}	2.1	2.0×10^{-8}	2.2	1.2×10^{-8}	2.8	1.2×10^{-8}	1.4	3.4×10^{-9}	4.9	3.6×10^{-9}	2.3	2.5×10^{-8}	2.5	1.9×10^{-8}	1.9

^a Corrected to 2/14/76.^b Standard deviation.^c NL - not calculated.

Table 5C-1. (Continued)

Sample ID Collection date Volume (l)	Cheshire B-5 5/28/85 184				Cheshire B-6 5/28/85 200				Blank Filterbags ^d 200			
	Summed fractions	% SD	Combined ashed	% SD	Summed fractions	% SD	Combined ashed	% SD	Summed fractions	% SD	Combined ashed	% SD
⁵⁴ Mn	$< 6.5 \times 10^{-10}$	—	7.0×10^{-9}	71	$< 3.0 \times 10^{-7}$	—	$< 2.0 \times 10^{-7}$	—	$< 2.3 \times 10^{-12}$	—	$< 9.0 \times 10^{-13}$	—
⁶⁰ Co	7.8×10^{-11}	15.8	8.7×10^{-11}	4.4	5.3×10^{-11}	11.5	$< 7.9 \times 10^{-11}$	5.2	$< 2.7 \times 10^{-12}$	—	$< 9.0 \times 10^{-13}$	—
¹⁰⁶ Ru	1.5×10^{-6}	5.9	1.4×10^{-6}	2.2	1.1×10^{-6}	6.3	1.3×10^{-6}	3.9	$< 2.3 \times 10^{-11}$	—	$< 2.0 \times 10^{-12}$	—
¹²⁵ Sb	3.7×10^{-8}	1.6	3.6×10^{-8}	0.7	2.8×10^{-8}	1.6	3.2×10^{-8}	0.9	$< 6.3 \times 10^{-12}$	—	$< 2.0 \times 10^{-12}$	—
¹³⁷ Cs	6.0×10^{-8}	0.8	5.8×10^{-8}	0.8	4.6×10^{-8}	1.1	5.4×10^{-8}	1.0	$< 2.7 \times 10^{-12}$	—	$< 4.6 \times 10^{-12}$	—
¹⁴⁴ Ce	$< 8.8 \times 10^{-12}$	—	3.3×10^{-10}	31	$< 1.8 \times 10^{-11}$	—	$< 1.8 \times 10^{-11}$	—	$< 1.4 \times 10^{-11}$	—	$< 4.0 \times 10^{-11}$	—
¹⁵² Eu	1.1×10^{-10}	22.5	1.2×10^{-10}	4.8	8.0×10^{-11}	12.6	1.0×10^{-10}	8.1	$< 4.6 \times 10^{-12}$	—	$< 2.0 \times 10^{-12}$	—
¹⁵⁴ Eu	2.1×10^{-10}	7.4	2.1×10^{-10}	4.9	1.8×10^{-10}	14.2	2.0×10^{-10}	3.8	$< 7.7 \times 10^{-12}$	—	$< 9.0 \times 10^{-13}$	—
¹⁵⁵ Eu	5.8×10^{-10}	13.1	7.5×10^{-10}	4.9	4.8×10^{-10}	9.2	6.9×10^{-10}	7.3	1.0×10^{-11}	27	$< 2.0 \times 10^{-12}$	—

^d Activity at count time.

Table 5C-2. Ratios of combined ashed-composite to summed-filter activities.

Nuclide	B-1	(%) ^a	B-2	(%)	B-3	(%)	B-4	(%)	B-5	(%)	B-6	(%)	$\bar{x} \pm SD^b$
²⁴¹ Am	0.74 ^c	(0.27)	0.60 ^c	(0.32)	—	—	1.00 ^c	(0.80)	—	—	—	—	—
⁶⁰ Co	0.90	(0.01)	0.95	(0.03)	0.80	(0.05)	0.79	(0.03)	1.12 ^c	(0.18)	1.49 ^c	(0.18)	0.86 ± 0.08
¹⁰⁶ Ru	0.86	(0.03)	0.93	(0.05)	0.78	(0.07)	0.80	(0.04)	0.93	(0.06)	1.18	(0.09)	0.91 ± 0.15
¹²⁵ Sb	0.92	(0.02)	1.02	(0.04)	0.81	(0.02)	0.85	(0.02)	0.97	(0.02)	1.14	(0.02)	0.95 ± 0.12
¹³⁷ Cs	0.95	(0.01)	0.93	(0.01)	0.95	(0.01)	0.83	(0.01)	0.97	(0.01)	1.17	(0.02)	0.97 ± 0.11
¹⁴⁴ Ce	1.06	(0.11)	1.23 ^c	(0.34)	1.67 ^c	(0.36)	0.84 ^c	(0.15)	—	—	—	—	—
¹⁵² Eu	0.90	(0.02)	1.00	(0.04)	0.81	(0.05)	0.79	(0.02)	1.09 ^c	(0.25)	0.80	(0.02)	0.88 ± 0.10
¹⁵⁴ Eu	0.93	(0.02)	1.00	(0.03)	0.92	(0.04)	0.79	(0.02)	1.00	(0.09)	0.90 ^c	(0.13)	0.93 ± 0.09
¹⁵⁵ Eu	1.05	(0.03)	1.00	(0.03)	1.06	(0.06)	0.76	(0.02)	1.29 ^c	(0.18)	1.44 ^c	(0.17)	0.97 ± 0.14
$\bar{y} \pm SD$	0.95 ± 0.07		0.98 ± 0.34		0.88 ± 0.10		0.80 ± 0.03		0.97 ± 0.03		1.16 ± 0.02		0.92 ± 0.04

^a The % counting error for that nuclide in the sample.

^b Standard deviation.

^c Omitted from calculation of means.

Table 5C-3. Repeat count comparisons, ashed composite filters (units: $\mu\text{Ci/mL} \pm 1 \text{ SD}$).^a

Nuclide	B-5 ₁	(%) ^b	B-5 ₂	(%)	B-5 ₁ /B-5 ₂	B-6 ₁	(%)	B-6 ₂	(%)	B-6 ₁ /B-6 ₂
⁴⁰ K	5.61 × 10 ¹⁰	(5.0)	5.21 × 10 ¹⁰	(5.0)	1.08	4.30 × 10 ¹⁰	(7.9)	3.91 × 10 ¹⁰	(4.8)	1.09
⁶⁰ Co	8.68 × 10 ¹¹	(4.4)	7.21 × 10 ¹¹	(5.1)	1.20	7.86 × 10 ¹¹	(5.2)	8.10 × 10 ¹¹	(4.9)	0.97
¹⁰⁶ Ru	1.34 × 10 ⁶	(2.2)	1.36 × 10 ⁶	(3.5)	0.98	1.30 × 10 ⁶	(3.9)	1.26 × 10 ⁶	(5.3)	1.03
¹²⁵ Sb	3.61 × 10 ⁸	(0.7)	3.60 × 10 ⁸	(0.7)	1.00	3.24 × 10 ⁸	(0.9)	2.96 × 10 ⁸	(0.8)	1.09
¹³⁷ Cs	5.84 × 10 ⁸	(0.8)	5.67 × 10 ⁸	(1.1)	1.03	5.36 × 10 ⁸	(1.0)	4.94 × 10 ⁸	(0.7)	1.09
¹⁵² Eu	1.23 × 10 ¹⁰	(4.8)	1.16 × 10 ¹⁰	(5.4)	1.06	1.00 × 10 ¹⁰	(8.1)	9.03 × 10 ¹⁰	(5.0)	1.11
¹⁵⁴ Eu	2.06 × 10 ¹⁰	(4.9)	2.24 × 10 ¹⁰	(2.7)	0.92	1.99 × 10 ¹⁰	(3.8)	1.75 × 10 ¹⁰	(3.1)	1.14
¹⁵⁵ Eu	7.50 × 10 ¹⁰	(4.9)	7.99 × 10 ¹⁰	(5.2)	0.94	6.86 × 10 ¹⁰	(7.3)	5.20 × 10 ¹⁰	(5.0)	1.32
Mean (± SD) ratio					1.03 ± 0.09					1.11 ± 0.10

^a Standard deviation.

^b The % counting error for that nuclide in the sample.

(cesium, antimony, and ruthenium), so we can eliminate that consideration. Between-sample ratio comparisons suggest that some total loss did occur in the case of sample B-4, and this is supported by the laboratory records. This was the first sample ashed and the only sample that included flat Millipore filters with the bag filters that were subsequently used exclusively. On heating, these filters formed small pellets that could not be reduced to ash. We suspect that the difference is due to loss incurred in the extended efforts to ash these pellets or is a result of the altered geometry when we counted them.

Somewhat less obvious is the source of the apparent gain on ashed-composite sample B-6. Some, but not all, of this systematic difference could be removed if either the second ash count or the mean of the two counts had been used in the comparison (see Table 5C-3). We also note, however, that a majority of the high uncertainty ratios from all samples (compounded counting uncertainty >10% in Table 5C-2) have ratios greater than 1.0. The decision to exclude from the averages ratios with an uncertainty in excess of 10% was based on substantial semi-empirical experience; we have found that values with high calculated uncertainties are likely to present Compton and background correction problems and that the actual uncertainties are often greater than the calculated values. As can be seen from Table 5C-2, this group of ratios contains most of the extreme outliers. We have already noted that precision and sensitivity are superior for the ash samples, so these ratios are typically derived from a low-precision summed value and a significantly more precise ash value. This suggests that the values for the upper limits and barely detectable concentrations calculated by GAMANAL are systematically too low, since even the addition of the upper-limit values into the sums does not raise the values enough to bring the ash/sum ratio reasonably close to unity. We consider this possibility intriguing but not conclusive (the cesium and antimony values for B-6, for example, involve no limits or high uncertainties and still yield high ratios). On the whole, it tends to support the utility of the ashing process as an approach to improving sensitivity and precision.

The results presented in Table 5C-3 indicate that the actual precision, as determined by reproducibility, of the ash activity determinations is about $\pm 10\%$. This is consistent with our

general experience on the replicability of counting data, and also with the observed distribution of data summarized in Table 5C-2. This does not address the question of absolute accuracy, and it should be noted that the ashed-composite samples are probably compact and homogeneous relative to the bulk-filter samples in the tuna-can configuration.

Conclusions

The ashing procedure provides results that are consistent with the results obtained from a direct count of the filters. Although probably not justified for routine applications, ashing does provide increased sensitivity and precision in the determination of very low levels of particulate activity. Although the increased manipulation carries with it the potential for sample loss, this does not appear to be a significant problem with samples using the bag filters.

Comparison of the data sets suggests the following conclusions concerning the accuracy and precision of the gamma-ray counting data:

- Activities with GAMANAL-calculated counting uncertainties of 10% or more may be considerably less precise than the counting statistics indicate.
- There is the possibility that GAMANAL underestimates both the very-low-level activities and the upper limits it calculates.
- Although counting statistics appreciably less than 10% are required for a reliable determination, the actual reproducibility of the values obtained is probably best represented by an uncertainty of approximately 10%.

Section 5D. Gamma-Ray Spectrometry Intercalibration with LANL

Contributors: J. H. Rego, R. W. Buddemeier, and K. V. Marsh

Introduction

Both the Lawrence Livermore and the Los Alamos National Laboratories analyze large-volume, low-radioactivity groundwater samples from HRMP sites. In order to investigate possible biases occurring as a result of different processing and analysis methods, we exchanged samples from water collections at the Cheshire site. These were not duplicate samples

and were, in fact, processed by different techniques, but both samples were counted on both laboratories' systems and so are directly comparable on an individual sample basis. In each case, the selected sample had been filtered, evaporated to dryness, and the remaining salt packaged for counting in each laboratory's standard form.

LLNL Sample Preparation

The LLNL salt sample from Cheshire cavity sample B-3 (see Appendix A and Sec. 1) was homogenized and sealed in our standard "tuna-can" counting container, a geometry well-calibrated for our gamma-ray spectroscopy measurements. It was counted on four different Ge-Li spectrometers, and average values were calculated (see Table 5D-1). The standard deviation of the mean value was calculated without regard to statistical counting errors. The sample and our analytical results were sent to LANL. The results of the LANL measurements are reported by Thompson (1987); generally, all their measurements except for ^{40}K agreed with ours within 25% or better.

LANL Sample Preparation

LANL prepared the dried salt from their sample 852-9-85-004, which had been collected from the Cheshire formation (above the cavity) on 18 June 1985. Their standard procedure is to use hydraulic pressure to compact the sample into a plastic-bagged "hockey puck" configuration—a solid disk slightly smaller than the LLNL "tuna can." This sample was sent to LLNL along with two gamma-ray spectrometry printouts (one for a 500-minute "short" count and one for a 4000-minute "long" count) and the results of a radiochemical separation determination of ^{137}Cs .

LLNL Measurements and Results

On receiving the Los Alamos sample, we x-rayed it (top, bottom, and side views at 0 and 90 degrees) to determine packing uniformity. There were no large voids or asymmetries noted. The x-ray procedure was suggested because (as described below) the sample would be repackaged during our analysis: documentation of its original condition would aid in verifying any

counting discrepancies or in answering any questions regarding differences in geometry and void areas.

The LANL sample was then counted twice in the "hockey puck" configuration, the second time after rotation through 90 deg in the detection chamber, and the results of these two counts were averaged. The sample was then carefully removed from the plastic bags, broken up into particle sizes comparable to the original salt sample, repacked in one of our standard tuna cans, and counted again. Results of these counts, along with LANL's analyses, are shown in Table 5D-2. Table 5D-3 presents the ratios of the LANL activity to the LLNL activity, both for the LLNL determinations described and for the LANL determinations previously reported by Thompson (1987).

Discussion

In Sec. 5C, we discussed the fact that the reproducibility of the activity values using the same sample and calculation procedures is on the order of 10% for samples with calculated counting uncertainties significantly less than 10%, and is substantially greater for values with higher calculated uncertainties. The calculated counting uncertainty is typically lower than the actual experimental precision, and for relatively high counting uncertainties the calculated value may be a very poor estimate of the accuracy of the determination. This is due to a number of factors such as the large size of the sample and its lack of homogeneity, detector size (which affects peak/Compton ratios) and calibration, and counting geometry variations due to positioning errors from count to count. The interlaboratory comparison, although not conducted with "known" or independently determined activities, gives us at least some information about the accuracy as well as the precision of our determinations. Table 5D-1 presents the results of careful replicate counts on a relatively high-activity (cavity) sample; we consider the uncertainties in the absolute activities to represent the best precision that can be achieved, and we would normally consider ± 5 –10% to be a more conservative value.

Because the activity of the LANL-supplied formation sample was so much lower than the LLNL-supplied cavity sample analyzed by LANL, there are only three nuclides for which both

Table 5D-1. LLNL-measured nuclides in sample B3 (LANL intercomparison sample). ^a											
Nuclide	λ/yr	Det. AA ^b	(%) ^c	Det. VV ^b	(%)	Det. EE ^b	(%)	Det. R7 ^b	(%)	Average	(%) SD ^d
²² Na	0.269	6.27×10^{-9}	12.5	7.82×10^{-9}	11.6	8.21×10^{-9}	10	9.89×10^{-9}	13	8.05×10^{-9}	16
⁴⁰ K	5.4×10^{-10}	3.66×10^{-9}	8.5	3.25×10^{-9}	8.5	3.27×10^{-9}	5.7	3.82×10^{-9}	10.8	3.56×10^{-9}	6.6
⁶⁰ Co	0.132	2.94×10^{-9}	1.3	2.62×10^{-9}	1.6	2.95×10^{-9}	1.4	3.22×10^{-9}	1.6	2.98×10^{-9}	4.9
¹⁰⁶ Ru	0.690	1.14×10^{-5}	4	1.13×10^{-5}	5.2	1.12×10^{-5}	5	1.17×10^{-5}	4.9	1.14×10^{-5}	1.6
¹²⁵ Sb	0.250	6.60×10^{-6}	1.3	6.36×10^{-6}	0.6	6.73×10^{-6}	0.5	7.02×10^{-6}	0.7	6.68×10^{-6}	3.6
¹³⁴ Cs	0.340	8.08×10^{-9}	5.3	2.59×10^{-8}	6.5	2.72×10^{-8}	5.1	2.96×10^{-8}	7.3	2.27×10^{-8}	37.7
¹³⁷ Cs	0.023	4.55×10^{-6}	0.7	4.23×10^{-6}	0.7	4.46×10^{-6}	0.7	4.72×10^{-6}	0.8	4.49×10^{-6}	3.9
¹⁵² Eu	0.052	6.76×10^{-9}	2.4	6.52×10^{-9}	2.6	6.77×10^{-9}	2.1	7.14×10^{-9}	3.1	6.80×10^{-9}	3.2
¹⁵⁴ Eu	0.082	1.23×10^{-8}	2.3	1.14×10^{-8}	2.6	1.17×10^{-8}	2	1.22×10^{-8}	2.9	1.19×10^{-8}	3.1
¹⁵⁵ Eu	0.147	3.90×10^{-8}	2.1	3.33×10^{-8}	5	3.38×10^{-8}	3.2	3.57×10^{-8}	7.5	3.55×10^{-8}	6.3

^a Units in $\mu\text{Ci}/\text{ml}$, corrected for decay to 2/14/76.

^b Detector configurations: AA is a sidelooker; VV is a downlooker; and EE and R7 are uplookers.

^c The percent counting error for that nuclide in the sample.

^d Standard deviation on the mean of four counts.

^a Units in $\mu\text{Ci/mL}$, corrected for decay to 2/14/76.

^b Detector configurations: AA is a sideloader; VV is a downlooker; and EE and R7 are uplookers.

^c The percent counting error for that nuclide in the sample.

^d Standard deviation on the mean of four counts.

Table 5D-2. LANL and LLNL determinations of LANL intercomparison sample (units: $\mu\text{Ci/mL} \pm 1 \text{ SD}$).										
Nuclide	LANL determinations ^{a,b}				LLNL determinations					
	Short count	SD	Long count	SD	Radiochemistry	LANL form ^c	SD	LLNL form ^a	SD	SD
⁴⁰ K	3.8×10^{-9}	10.6	3.8×10^{-9}	3.9	—	2.0×10^{-9}	0.8	2.4×10^{-9}	3.1	—
⁶⁰ Co	—	—	1.1×10^{-10}	17.2	—	2.7×10^{-12}	—	$<1.7 \times 10^{-11}$	—	—
¹⁰⁶ Ru	2.5×10^{-8}	14.9	1.1×10^{-6}	45.0	—	8.2×10^{-7}	19.3	1.6×10^{-6}	19.2	—
¹²⁵ Sb	3.0×10^{-6}	4.9	3.0×10^{-6}	2.5	—	2.9×10^{-6}	2.8	4.0×10^{-6}	0.5	—
¹³⁷ Cs	4.7×10^{-7}	0.5	—	—	7.1×10^{-7}	4.9×10^{-7}	0.1	6.3×10^{-7}	0.7	—
¹⁵² Eu	5.5×10^{-8}	36.3	4.2×10^{-11}	8.0	—	5.7×10^{-11}	67.5	$<1.0 \times 10^{-11}$	—	—
¹⁵⁴ Eu ^d	6.7×10^{-10}	21.9	—	—	—	1.5×10^{-10}	31.7	$<1.5 \times 10^{-11}$	—	—
¹⁵⁵ Eu	—	—	5.9×10^{-9}	36.3	—	8.4×10^{-10}	13.1	$<8.3 \times 10^{-11}$	—	—

^a Uncertainties based on counting statistics.

^b LANL form; long-count value used for comparisons if available.

^c Uncertainties based on mean if two replicate counts.

^d LANL determination questionable.

Table 5D-3. Ratio of LANL counts in LANL form to LLNL counts.

Nuclide	LANL determination of B-3		LLNL determination ^a of 852-9-85-004	
	RAYGUN code	SPECANL code	LANL configuration	LLNL configuration
⁴⁰ K	1.42	1.48	1.90	1.58
⁶⁰ Co	1.09	1.04	—	—
¹⁰⁶ Ru	1.00	1.21	1.34	0.69
¹²⁵ Sb	0.85	0.80	1.03	0.75
¹³⁴ Cs	0.93	1.01	—	—
¹³⁷ Cs	0.75	0.75	0.96	0.75
			1.45 ^b	1.13 ^b
¹⁵² Eu	0.93	0.89	0.74	—
¹⁵⁴ Eu	1.15	1.12	4.47	—
¹⁵⁵ Eu	1.15	1.18	7.02	—

^a "Long count" value from Table 5D-1 used for ratios, if available.

^b Relative to radiochemical separation value in Table 5D-1.

the LANL and the LLNL analyses achieved counting statistics less than 10%— ^{40}K , ^{125}Sb , and ^{137}Cs (see Table 5D-2). If we compare the activity ratios for these nuclides (LANL count in LANL form to LLNL count in LLNL form) for the two different sample intercomparisons in Table 5D-2, we see that they are quite similar. At the level of upper-limit and/or highly imprecise determinations, agreement is no better than to within an order of magnitude. The results do not contain any indication of which set of numbers is more accurate, although the radiochemical ^{137}Cs result is somewhat closer to the LLNL gamma-ray spectrometry value.

Although the LLNL intercomparison set is too small to be treated as statistically significant, most of the ratios that are not based on imprecise determinations (and some that are) deviate from unity by 25–35%—similar to the 25% range noted for the LANL intercomparison set. We may ask what sort of distribution of ratios we would expect to see as a function of the uncertainties of the individual numbers (considering accuracy as well as precision). If two numbers both have an associated (one standard deviation) uncertainty of 25%, then their ratio will have an uncertainty of 35%; the comparable ratio uncertainties for component number uncertainties of 20, 15, and 10% are 28, 21, and 14%. Although the ratio consistency for individual nuclides may suggest that we are dealing with biases rather than random errors, this analysis indicates that, in the absence of better data, an appropriate conservative value to assign as the actual uncertainty of an activity value with counting statistics <10% is probably in the range of 20 to 25%.

Conclusions

The LANL-LLNL intercomparison efforts indicate that the two laboratories agree to within 25% on the activities of nuclides with counting uncertainties <10%, although the reproducibility of individual numbers by an individual laboratory may be considerably better. There is a suggestion that some of the differences between the LANL and LLNL determinations for individual nuclides may represent systematic biases rather than random errors; however, pending further calibration results, we suggest that a conservative approach to data interpretation is to assign an absolute uncertainty of $\pm 25\%$ to all gamma-ray spectrometry results based

on counting statistics $<10\%$, and to treat all values with higher uncertainties as order-of-magnitude estimates. If this is done, the LANL and LLNL results will generally be in complete agreement for the same sample, and differences beyond these levels of uncertainty can normally be ascribed to the sample rather than to the techniques.

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**Appendix A. Analytical Data
Used in Study of Radionuclide
Migration, Cheshire Area**

Table A-1. Cheshire (U20n) sample B-1 nuclide analyses.^a

Isotope	Fraction ID $t_{1/2}$ (yr)	Prefilter		0.45- μm filter		0.20- μm filter		Ashed com- posite filters		Retentate >0.006 μm		Filtrate (salt) <0.006 μm	
		Value	%SD	Value	%SD	Value	%SD	Value	%SD	Value	%SD	Value	%SD
¹ H	12.26	—	—	—	—	—	—	—	—	—	—	0.615	1.0
²³ Na	2.605	$<9.6 \times 10^{13}$	—	4.63×10^{10}	37.8	$<9.6 \times 10^{13}$	—	3.35×10^{10}	70.5	$<4.5 \times 10^{11}$	—	7.11×10^9	3.8
⁴⁰ K	1.3×10^9	$<4.8 \times 10^{11}$	—	$<4.1 \times 10^{11}$	—	$<3.9 \times 10^{11}$	—	3.60×10^{10}	14.8	1.67×10^9	19.7	1.00×10^9	18.9
⁵⁴ Mn	0.855	5.92×10^8	8.5	8.11×10^9	21.1	6.88×10^{-9}	31	5.77×10^8	33.8	4.34×10^8	57	$<1.6 \times 10^{11}$	—
⁶⁰ Co	5.272	3.18×10^9	0.9	4.56×10^{10}	2.1	3.93×10^{-10}	1.9	3.61×10^{-9}	0.7	1.50×10^9	3	$<6.7 \times 10^{12}$	—
¹⁰⁶ Ru	1.020	2.37×10^6	2	1.05×10^6	2.6	8.34×10^{-7}	2.8	3.73×10^6	2.5	7.25×10^6	3.2	1.03×10^6	26.4
¹²⁵ Sb	2.760	3.89×10^8	2.2	1.58×10^8	3	1.14×10^{-8}	1.3	6.07×10^8	1.5	2.07×10^7	2.6	5.00×10^6	1.2
¹³⁴ Cs	2.065	1.19×10^9	20.5	4.37×10^{10}	3.9	3.47×10^{-10}	14.5	1.87×10^9	9.7	7.02×10^9	9.3	1.28×10^8	3.6
¹³⁷ Cs	30.170	2.68×10^7	0.8	7.57×10^{-8}	0.9	5.11×10^{-8}	0.8	3.72×10^7	0.8	1.04×10^6	1.1	1.76×10^6	0.8
¹⁴⁴ Ce	284	3.37×10^9	6.4	8.86×10^{10}	10.6	6.57×10^{-10}	11	5.20×10^9	7.1	7.08×10^9	9.2	ND ^b	—
¹⁵² Eu	13.400	2.67×10^9	1.7	6.73×10^{10}	2.5	4.98×10^{-10}	2	3.54×10^{-9}	1.2	5.67×10^9	2.8	$<3.2 \times 10^{11}$	—
¹⁵⁴ Eu	8.500	4.69×10^9	1.2	1.19×10^9	2.2	8.77×10^{-10}	1.4	6.18×10^{-9}	1.2	9.64×10^9	2	$<7.2 \times 10^{11}$	—
¹⁵⁵ Eu	4.730	1.28×10^8	1.7	3.34×10^9	2.9	2.42×10^{-9}	2.9	2.00×10^8	2.2	2.24×10^8	5.4	$<1.5 \times 10^{10}$	—

^a t_0 = detonation time (day 45, 1976). Values are in $\mu\text{Ci/mL} \pm 1$ SD (%) at t_0 . Date collected: 9/8/83; sample volume: 188 L; pumped volume: 56,790 gal; location: cavity. Detection limits are calculated at count time. Evaporation after filtration through 0.006- μm UF. Filtered 364 days after collection. Sample recycled through entire filter series during ultrafiltration.

^b Not detected. No limit calculated for this nuclide.

Table A-2. Cheshire (U20n) sample B-2 nuclide analyses.^a

Isotope	Fraction ID	$t_{1/2}$ (yr)	Prefilter		0.45- μ m filter		Ashed com- posite filters		Filtrate (salt ^b) < 0.20 μ m	
			Value	%SD	Value	%SD	filters	%SD	Value	%SD
¹ H		12.33	—	—	—	—	—	—	0.614	1.0
²² Na		2.605	4.71×10^{-10}	37.8	3.04×10^{-10}	36.2	2.29×10^{-10}	75.9	9.01×10^{-9}	8.7
⁴⁰ K		1.3×10^9	1.41×10^{-9}	11.6	ND ^c	—	1.22×10^{-9}	6.1	3.83×10^{-9}	6.8
⁵⁴ Mn		0.855	3.01×10^{-8}	21.1	1.09×10^{-8}	31.4	2.89×10^{-8}	46.4	7.93×10^{-8}	32.1
⁶⁰ Co		5.272	1.52×10^{-9}	2.1	4.96×10^{-10}	4.3	1.93×10^{-9}	0.9	4.05×10^{-9}	1.3
¹⁰⁶ Ru		1.020	2.31×10^{-6}	2.6	4.35×10^{-7}	4.9	2.46×10^{-6}	4.8	1.21×10^{-5}	1.7
¹²⁵ Sb		2.760	3.65×10^{-8}	3	7.65×10^{-9}	3.4	4.47×10^{-8}	1.7	6.23×10^{-6}	0.8
¹³⁴ Cs		2.065	4.66×10^{-9}	3.9	2.20×10^{-10}	41.4	4.32×10^{-9}	4.4	2.19×10^{-8}	5.2
¹³⁷ Cs		30.170	6.76×10^{-7}	0.9	4.38×10^{-6}	0.7	6.74×10^{-7}	1.1	2.68×10^{-6}	0.7
¹⁴⁴ Ce		284	2.65×10^{-9}	10.6	4.69×10^{-10}	11	3.79×10^{-9}	25.6	1.18×10^{-8}	6.1
¹⁵² Eu		13.400	2.05×10^{-9}	2.5	3.14×10^{-10}	11.3	2.38×10^{-9}	1.3	9.64×10^{-9}	2.1
¹⁵⁴ Eu		8.500	2.75×10^{-9}	2.2	4.58×10^{-10}	9.5	4.22×10^{-9}	1.1	1.58×10^{-8}	2.1
¹⁵⁵ Eu		4.730	9.87×10^{-9}	2.9	1.75×10^{-9}	2	1.24×10^{-9}	1.4	4.94×10^{-8}	2.5

^a t_0 = detonation time (day 45, 1976). Values are in μ Ci/mL $\pm$ 1 SD (%) at t_0 . Date collected: 9/9/83; sample volume: 200 L; pumped volume: 131,860 gal; location: cavity. Detection limits are calculated at count time. Filtered 87 days after collection.

^b Evaporation after filtration through 0.45- μ m filter.

^c Not detected. No limit calculated for this nuclide.

Table A-3. Cheshire (U20n) sample B-3 nuclide analyses.^a

Isotope	Fraction ID	$t_{1/2}$ (yr)	Prefilter		0.45- μ m filter		0.20- μ m filter		Ashed composite filters		(salt) < 0.20 μ m	
			Value	%SD	Value	%SD	Value	%SD	filters	%SD	Value	%SD
³ H		12.33	—	—	—	—	—	—	—	—	0.504	1.0
²² Na		2.605	1.55×10^{10}	37.3	$< 6.0 \times 10^{12}$	—	$< 1.1 \times 10^{12}$	—	2.32×10^{10}	31.6	7.87×10^9	16
⁴⁰ K		1.3×10^9	$< 4.9 \times 10^{11}$	—	$< 2.2 \times 10^{10}$	—	$< 6.1 \times 10^{11}$	—	1.19×10^{10}	20.6	3.46×10^9	6.6
⁵⁴ Mn		0.855	$< 7.6 \times 10^{12}$	—	$< 4.6 \times 10^{12}$	—	$< 2.2 \times 10^{12}$	—	1.23×10^8	64.3	8.40×10^8	72.3
⁶⁰ Co		5.272	2.33×10^{10}	3.8	1.20×10^{10}	17.3	1.05×10^{10}	16.9	3.75×10^{10}	1.5	2.96×10^9	4.89
¹⁰⁶ Ru		1.020	3.37×10^7	6.8	3.32×10^{-7}	10	2.80×10^{-7}	7.9	7.45×10^7	3.4	1.05×10^5	1.58
¹²⁵ Sb		2.760	7.40×10^{-9}	2.2	1.05×10^{-6}	2.3	8.40×10^{-9}	2.1	2.14×10^8	1.6	6.74×10^6	3.58
¹³⁴ Cs		2.065	2.29×10^{10}	20.1	ND ^b	—	ND	—	4.27×10^{10}	19.2	2.68×10^8	37.6
¹³⁷ Cs		30.170	3.99×10^8	0.8	3.18×10^8	0.9	3.34×10^{-8}	1	9.47×10^6	0.8	4.49×10^6	3.92
¹⁴⁴ Ce		284	5.12×10^{10}	17.5	$< 4.5 \times 10^{11}$	—	$< 2.9 \times 10^{11}$	—	8.49×10^{10}	12.5	9.54×10^9	15.2
¹⁵² Eu		13.400	3.55×10^{10}	4.3	1.81×10^{10}	10.3	2.02×10^{10}	4.8	6.01×10^{10}	2.4	6.78×10^9	3.22
¹⁵⁴ Eu		8.500	5.79×10^{10}	3.4	3.28×10^{10}	6.2	3.37×10^{10}	2.9	1.10×10^9	1.8	1.19×10^8	3.1
¹⁵⁵ Eu		4.730	1.68×10^{-9}	3.7	8.59×10^{-10}	7.9	9.08×10^{-10}	4.3	3.63×10^{-9}	2.3	3.53×10^8	6.3

^a t_0 = detonation time (day 45, 1976). Values are in μ Ci/mL $\pm$ 1 SD (%) at t_0 . Date collected: 10/23/84; sample volume: 166 L; pumped volume: 3.0×10^6 gal; location: cavity. Detection limits are calculated at count time. Evaporation after filtration through 0.20- μ m filter. Filtered 134 days after collection.

^b Not detected. No limit calculated for this nuclide.

Table A-4. Cheshire (U20n) sample B-4 nuclide analyses.^a

Isotope	Fraction ID	$t_{1/2}$ (yr)	Prefilter ^b		0.45- μ m filter		0.20- μ m filter		0.05- μ m filter	
			(Sum)	%SD	Value	%SD	(Sum)	%SD	Value	%SD
³ H		12.33	—	—	—	—	—	—	—	—
²² Na		2.605	$<1.3 \times 10^{-10}$	—	$<9.3 \times 10^{-12}$	—	1.88×10^{-10}	—	1.11×10^{-10}	87.5
⁴⁰ K		1.3×10^9	8.23×10^{-10}	16.1	1.61×10^{-10}	27.4	3.26×10^{-10}	24	1.37×10^{-9}	4.5
⁵⁴ Mn		0.855	2.48×10^{-8}	45.8	8.26×10^{-9}	44.9	$<2.7 \times 10^{-12}$	—	$<2.3 \times 10^{-12}$	—
⁶⁰ Co		5.272	1.15×10^{-9}	1.8	4.01×10^{-10}	7.7	4.82×10^{-10}	2.1	3.82×10^{-10}	6.4
¹⁰⁶ Ru		1.020	3.39×10^{-6}	2.8	1.36×10^{-6}	7.7	1.66×10^{-6}	3.5	7.32×10^{-7}	8
¹²⁵ Sb		2.760	4.70×10^{-8}	2.1	3.33×10^{-8}	2.3	2.62×10^{-8}	2.1	2.41×10^{-8}	2.8
¹³⁴ Cs		2.065	2.97×10^{-9}	7.3	1.00×10^{-9}	16.8	1.29×10^{-9}	9.8	1.79×10^{-9}	21
¹³⁷ Cs		30.170	5.11×10^{-7}	1.1	1.78×10^{-7}	1.1	2.40×10^{-7}	0.8	3.00×10^{-7}	1
¹⁴⁴ Ce		284	3.46×10^{-9}	12.4	1.26×10^{-9}	34.2	1.34×10^{-9}	13.5	6.75×10^{-10}	4.2
¹⁵² Eu		13.400	2.42×10^{-9}	1.8	8.59×10^{-10}	4.2	1.09×10^{-9}	2	9.12×10^{-10}	3.5
¹⁵⁴ Eu		8.500	4.24×10^{-9}	1.2	1.51×10^{-9}	2.6	1.88×10^{-9}	1.6	1.48×10^{-9}	5
¹⁵⁵ Eu		4.730	1.19×10^{-8}	1.7	4.08×10^{-9}	4.3	5.11×10^{-9}	2.1	3.75×10^{-9}	3.8

^a t_0 = detonation time (day 45, 1976). Values are in μ Ci/mL $\pm$ 1 SD (%) at t_0 . Date collected: 10/23/84; sample volume: 194.5 L; pumped volume: 3.0×10^6 gal; location: cavity. Evaporation after filtration to 0.003 μ m. Filtered 193 days after collection. Detection limits are calculated at count time.

^b Possible prefilter bag split.

Table A-4. (Continued)

Isotope	Fraction ID	$t_{1/2}$ (yr)	Ashed com- posite filters		Retentate >0.003 μm		Recount retentate >0.003 μm		Filtrate (salt) ^a <0.003 μm	
			Value	%SD	Value	%SD	Value	%SD	Value	%SD
³ H		12.33	—	—	—	—	—	—	0.504	1.0
²² Na		2.605	6.95×10^{-10}	38.2	3.18×10^{-10}	69	—	—	7.91×10^{-9}	3.3
⁴⁰ K		1.3×10^9	2.12×10^{-9}	3.6	3.50×10^{-10}	24.2	5.17×10^{-10}	8.0	1.12×10^{-9}	8.1
⁵⁴ Mn		0.855	3.33×10^{-8}	67.2	ND ^b	—	—	—	$<2.0 \times 10^{-11}$	—
⁶⁰ Co		5.272	1.94×10^{-9}	0.5	6.37×10^{-10}	6.3	6.95×10^{-10}	3	6.40×10^{-11}	30
¹⁰⁶ Ru		1.020	5.71×10^{-6}	2.9	1.87×10^{-6}	7.6	2.34×10^{-6}	7.8	1.52×10^{-6}	24.4
¹²⁵ Sb		2.760	1.06×10^{-7}	1.2	1.03×10^{-7}	1.1	1.21×10^{-7}	1	7.07×10^{-6}	9.4
¹³⁴ Cs		2.065	5.40×10^{-9}	4.5	1.53×10^{-9}	14.6	2.06×10^{-9}	13.4	1.81×10^{-8}	5
¹³⁷ Cs		30.170	1.02×10^{-6}	0.7	3.28×10^{-7}	0.8	3.78×10^{-7}	0.7	2.83×10^{-6}	0.7
¹⁴⁴ Ce		284	5.57×10^{-9}	8.5	2.82×10^{-9}	19.7	—	—	$<5.1 \times 10^{-10}$	—
¹⁵² Eu		13.400	4.16×10^{-9}	1.2	1.41×10^{-9}	3.4	1.68×10^{-9}	1.8	5.52×10^{-11}	60.1
¹⁵⁴ Eu		8.500	7.22×10^{-9}	1	2.54×10^{-9}	3.1	3.02×10^{-9}	1.3	$<7.6 \times 10^{-11}$	—
¹⁵⁵ Eu		4.730	1.93×10^{-8}	1.9	9.67×10^{-9}	2.8	9.21×10^{-9}	3.8	$<2.9 \times 10^{-10}$	—

^a Evaporation after ultrafiltration to 0.003 μm .^b Not detected. No limit calculated for this nuclide.

Table A-5. Cheshire (U20n) sample B-5 nuclide analyses.^a

Isotope	Fraction ID	$t_{1/2}$ (yr)	Prefilter ^b		0.45- μ m filter		0.20- μ m filter		0.05- μ m filter	
			(Sum)	%SD	Value	%SD	(Sum)	%SD	Value	%SD
³ H		12.26	—	—	—	—	—	—	—	—
²² Na		2.605	<2.1 × 10 ⁻¹²	—	<2.0 × 10 ⁻¹²	—	6.13 × 10 ⁻¹¹	84.8	<2.4 × 10 ⁻¹¹	—
⁴⁰ K		1.3 × 10 ⁹	1.34 × 10 ⁻¹⁰	23.6	<3.4 × 10 ⁻¹¹	—	<3.3 × 10 ⁻¹¹	—	3.48 × 10 ⁻¹⁰	16.2
⁵⁴ Mn		0.855	<1.4 × 10 ⁻¹²	—	<1.2 × 10 ⁻¹²	—	<1.5 × 10 ⁻¹²	—	<1.7 × 10 ⁻¹²	—
⁶⁰ Co		5.272	4.66 × 10 ⁻¹¹	12	<1.5 × 10 ⁻¹²	—	1.03 × 10 ⁻¹¹	8.8	3.11 × 10 ⁻¹¹	23.8
¹⁰⁶ Ru		1.020	4.37 × 10 ⁻⁷	1.2	1.72 × 10 ⁻⁷	20	1.29 × 10 ⁻⁷	9.3	7.69 × 10 ⁻⁷	4.5
¹²⁵ Sb		2.760	9.67 × 10 ⁻⁹	5.5	6.40 × 10 ⁻⁹	1.4	5.44 × 10 ⁻⁹	2.7	1.52 × 10 ⁻⁸	1.6
¹³⁴ Cs		2.065	ND ^c	—	ND	—	ND	—	ND	—
¹³⁷ Cs		30.170	1.36 × 10 ⁻⁸	0.8	3.95 × 10 ⁻⁹	0.9	5.56 × 10 ⁻⁹	0.8	3.68 × 10 ⁻⁸	0.7
¹⁴⁴ Ce		284	<1.4 × 10 ⁻¹¹	—	<8.3 × 10 ⁻¹²	—	<9.8 × 10 ⁻¹²	—	<2.2 × 10 ⁻¹¹	—
¹⁵² Eu		13.400	3.89 × 10 ⁻¹¹	30.7	<6.6 × 10 ⁻¹²	—	<7.6 × 10 ⁻¹²	—	7.55 × 10 ⁻¹¹	18.3
¹⁵⁴ Eu		8.500	7.33 × 10 ⁻¹¹	19.2	<2.5 × 10 ⁻¹²	—	<3.4 × 10 ⁻¹²	—	1.39 × 10 ⁻¹⁰	1.2
¹⁵⁵ Eu		4.730	1.52 × 10 ⁻¹⁰	18.2	3.00 × 10 ⁻¹¹	43.2	5.81 × 10 ⁻¹¹	0.8	3.36 × 10 ⁻¹⁰	10.2

^a t_0 = detonation time (day 45, 1976). Values are in μ Cl/mL $\pm$ 1 SD (%) at t_0 . Date collected: 5/28/85; sample volume: 184 L; pumped volume: 3.7×10^6 gal; location: outside cavity. Detection limits are calculated at count time. Filtered 85 days after collection.

^b Possible prefilter bag split.

^c Not detected. No limit calculated for this nuclide.

Table A-5. (Continued)										
Isotope	Fraction ID	$t_{1/2}$ (yr)	Ashed com- posite filters		Ashed com- posite filters		Retentate >0.003 μm		Filtrate (salt) ^d <0.003 μm	
			Value	%SD	Recount	%SD	Value	%SD	Value	%SD
³ H		12.33	—	—	—	—	—	—	0.433	1.0
²² Na		2.605	5.96×10^{-11}	54.6	—	—	$<1.7 \times 10^{-11}$	—	1.01×10^{-9}	5.8
⁴⁰ K		1.3×10^9	5.61×10^{-10}	5.0	5.21×10^{-10}	5	2.45×10^{-10}	9.4	2.21×10^{-9}	2
⁵⁴ Mn		0.855	7.00×10^{-9}	71.2	—	—	$<7.3 \times 10^{-13}$	—	$<2.0 \times 10^{-12}$	—
⁶⁰ Co		5.272	8.68×10^{-11}	4.4	7.21×10^{-11}	5.1	1.78×10^{-11}	16	$<1.2 \times 10^{-12}$	—
¹⁰⁶ Ru		1.020	1.37×10^{-6}	2.2	1.34×10^{-6}	3.9	5.13×10^{-7}	4.4	6.52×10^{-7}	17.2
¹²⁵ Sb		2.760	3.61×10^{-8}	0.7	3.60×10^{-8}	0.7	2.37×10^{-8}	0.6	3.87×10^{-6}	0.6
¹³⁴ Cs		2.065	5.75×10^{-11}	76.6	6.18×10^{-11}	9.4	4.51×10^{-11}	61	1.86×10^{-9}	7.7
¹³⁷ Cs		30.170	5.84×10^{-8}	0.8	5.67×10^{-8}	1.1	2.63×10^{-8}	0.7	6.57×10^{-7}	0.7
¹⁴⁴ Ce		284	3.35×10^{-10}	30.9	—	—	$<1.2 \times 10^{-11}$	—	$<7.5 \times 10^{-11}$	—
¹⁵² Eu		13.400	1.23×10^{-10}	4.8	1.16×10^{-10}	5.4	5.03×10^{-11}	7.4	$<5.6 \times 10^{-12}$	—
¹⁵⁴ Eu		8.500	2.06×10^{-10}	4.9	2.24×10^{-10}	2.7	8.63×10^{-11}	4.5	$<1.0 \times 10^{-11}$	—
¹⁵⁵ E ⁻⁴		4.730	7.50×10^{-10}	4.9	7.99×10^{-10}	5.2	3.04×10^{-10}	8.2	$<2.7 \times 10^{-11}$	—
^d Evaporation after ultrafiltration to 0.003 μm .										

Table A-6. Cheshire (U20n) sample B-6 nuclide analyses. ^a										
Isotope	Fraction ID	$t_{1/2}$ (yr)	Prefilter ^b		0.45- μ m filter		0.20- μ m filter		0.05- μ m filter	
			(Sum)	%SD	Value	%SD	(Sum)	%SD	Value	%SD
³ H		12.33	—	—	—	—	—	—	—	—
²³ Na		2.605	3.36×10^{10}	18.0	$<1.5 \times 10^{-12}$	—	1.32×10^{10}	72.2	$<1.1 \times 10^{12}$	—
⁴⁰ K		1.3×10^9	1.01×10^{10}	31.3	$<4.0 \times 10^{-11}$	—	$<2.6 \times 10^{11}$	—	2.61×10^{10}	11.1
⁵⁴ Mn		0.855	$<1.4 \times 10^{12}$	—	$<1.3 \times 10^{-12}$	—	$<1.4 \times 10^{11}$	—	$<6.8 \times 10^{13}$	—
⁶⁰ Co		5.272	5.26×10^{11}	11.5	$<1.5 \times 10^{-12}$	—	$<1.4 \times 10^{12}$	—	$<1.4 \times 10^{12}$	—
¹⁰⁶ Ru		1.020	5.23×10^7	4.6	1.50×10^{-7}	10.9	7.04×10^8	19.1	3.89×10^7	4.6
¹²⁵ Sb		2.760	1.06×10^8	1.4	4.08×10^{-9}	1.8	3.74×10^9	2.9	9.26×10^9	1.2
¹³⁴ Cs		2.065	ND ^c	—	ND	—	ND	—	ND	—
¹³⁷ Cs		30.170	1.56×10^8	1.6	3.86×10^{-9}	1.0	2.82×10^9	0.8	2.42×10^8	0.8
¹⁴⁴ Ce		284	$<1.3 \times 10^{11}$	—	$<8.9 \times 10^{-12}$	—	$<7.2 \times 10^{12}$	—	$<1.3 \times 10^{11}$	—
¹⁵² Eu		13.400	3.63×10^{11}	16.4	$<5.6 \times 10^{-12}$	—	$<5.2 \times 10^{12}$	—	4.42×10^{11}	9.4
¹⁵⁴ Eu		8.500	8.69×10^{11}	16.4	2.30×10^{-11}	25.1	$<2.0 \times 10^{12}$	—	6.56×10^{11}	7.4
¹⁵⁵ Eu		4.730	1.92×10^{10}	13.2	$<5.0 \times 10^{-12}$	—	$<3.2 \times 10^{12}$	—	2.81×10^{10}	6.5

^a t_0 = detonation time (day 45, 1976). Values are in μ Ci/mL $\pm$ 1 SD (%) at t_0 . Date collected: 5/28/85; sample volume: 200 L; pumped volume: 3.7×10^6 gal; location: outside cavity. Detection limits are calculated at count time. Filtered 181 days after collection.

^b Possible prefilter bag split.

^c Not detected. No limit calculated for this nuclide.

Table A-6. (Continued)										
Isotope	Fraction ID	$t_{1/2}$ (yr)	Ashed com- posite filters		Ashed com- posite filters		Retentate >0.003 μm		Filtrate (salt ^d) <0.003 μm	
			Value	%SD	Recount	%SD	Value	%SD	Value	%SD
³ H		12.33	—	—	—	—	—	—	0.433	1.0
²² Na		2.605	<7.1 × 10 ⁻¹¹	—	<7.1 × 10 ⁻¹¹	—	<9.0 × 10 ⁻¹³	—	1.02 × 10 ⁻⁹	4.7
⁴⁰ K		1.3 × 10 ⁹	4.30 × 10 ⁻¹⁰	7.9	3.94 × 10 ⁻¹⁰	4.8	5.61 × 10 ⁻¹¹	15.4	2.37 × 10 ⁻⁹	2.2
⁵⁴ Mn		0.855	<9.0 × 10 ⁻¹³	—	—	—	<4.5 × 10 ⁻¹³	—	<2.2 × 10 ⁻¹²	—
⁶⁰ Co		5.272	7.86 × 10 ⁻¹¹	5.2	8.10 × 10 ⁻¹¹	4.9	<6.8 × 10 ⁻¹³	—	<1.3 × 10 ⁻¹²	—
¹⁰⁶ Ru		1.020	1.30 × 10 ⁻⁶	3.9	1.26 × 10 ⁻⁶	5.3	9.56 × 10 ⁻⁸	11.4	5.66 × 10 ⁻⁷	24.3
¹²⁵ Sb		2.760	3.24 × 10 ⁻⁸	0.9	2.96 × 10 ⁻⁸	0.8	6.88 × 10 ⁻⁹	1.2	3.95 × 10 ⁻⁶	0.5
¹³⁴ Cs		2.065	ND	—	—	—	ND	—	3.52 × 10 ⁻¹⁰	25.7
¹³⁷ Cs		30.170	5.36 × 10 ⁻⁸	1	4.94 × 10 ⁻⁶	0.7	5.45 × 10 ⁻⁹	0.7	4.94 × 10 ⁻⁷	0.7
¹⁴⁴ Ce		284	<1.6 × 10 ⁻¹¹	—	—	—	<4.3 × 10 ⁻¹²	—	<9.4 × 10 ⁻¹¹	—
¹⁵² Eu		13.400	1.00 × 10 ⁻¹⁰	8.1	9.03 × 10 ⁻¹¹	5	9.30 × 10 ⁻¹²	26.3	<1.3 × 10 ⁻¹¹	—
¹⁵⁴ Eu		8.500	1.99 × 10 ⁻¹⁰	3.8	1.75 × 10 ⁻¹⁰	3.1	1.63 × 10 ⁻¹¹	11.9	<1.0 × 10 ⁻¹¹	—
¹⁵⁵ Eu		4.730	6.86 × 10 ⁻¹⁰	7.3	5.20 × 10 ⁻¹⁰	5	5.05 × 10 ⁻¹¹	20.3	<4.8 × 10 ⁻¹¹	—
^d Evaporation after ultrafiltration to 0.303 μm .										

**Appendix B. Soil Moisture Data
and Environmental Tritium
and Deuterium Analyses,
Cambria Area**

Key to Abbreviations used in Appendix B Tables

AMS: Air-moisture sample. Surface-air moisture was collected using silica-gel traps; one sampling was within the ditch vegetation and one was 92 ft south (upwind) of the ditch. Both were in the vicinity of Flume 1; both had air intakes about 3 ft above the surface. The first of the two numbers for each location represents a daytime sample; the second, a nighttime sample. Silica-gel samples were freeze dried at LLNL, with tritium analysis done at LLNL and deuterium analysis done at the University of Nevada, Las Vegas, Nevada. See Fig. 3A-1.

CCW: Cambrian canal water. Grab samples were collected at several locations along the ditch during the course of the study.

CPW: Cambrian pond water. Two surface-water sample surveys were conducted to identify temporal and spatial variation of tritium within the discharge pond and between the ditch and pond. The numerical suffixes in the W-series samples correspond to the locations mapped in Fig. 3A-1; other location designators are defined below.

EEOL: East end of lake. Several grab samples were collected along the eastern edge of the pond (lake) prior to the pond-sampling surveys.

F: Flume. Three calibrated Parshall flumes were installed in the ditch in order to estimate flow rates, percolations, and evaporative rates. Ditch-water grab samples were collected at each of these three locations over time. See Fig. 3A-1.

FDPW: Freeze-dried plant water. Plant material was sampled and freeze dried, and the extracted water was analyzed for tritium and deuterium. For the samples collected on 83271, the numbers after the "/" in the code column are the DRI TPS (see Table B3 for numbers that are field-collection replicates of the LLNL sample). For the samples collected on 84220, SC = salt cedar and CT = cattail; numbers after the CT designator indicate the height in feet above the water surface from which the reed sample was collected.

FDSW: Freeze-dried soil water. Soil-core water samples were extracted by freeze drying for tritium analysis. Location codes: for codes of the form 5-10, the first number is the distance in feet from the edge of the ditch berm; the second number is the depth in feet of the sample below ground surface. All were taken in or near the instrumented plot. The FF sample series was submitted by DRI. All samples of the form L-100-15 were from the L-100 installation borehole (see Fig. 3A-1 and 3A-5), with the final number indicating depth.

Isotopic concentration units:

Deuterium:

$$\%SMOW = [D/H(\text{sample})]/[D/H(\text{std})] \times 100$$

$$D(\%SMOW) = [1 - (D/H(\text{sample})/[D/H(\text{std})])] \times 1000$$

Tritium:

$$1 \text{ dpm/mL} = 4.50 \times 10^{-1} \text{ pCi/mL}$$

$$= 4.50 \times 10^{-7} \mu\text{Ci/mL} = 1.4 \times 10^{-2} \text{ T.U.}$$

$$1 \text{ pCi/mL} = 10^{-6} \mu\text{Ci/mL}$$

$$= 2.22 \text{ dpm/mL} = 3.10 \times 10^2 \text{ T.U.}$$

$$1 \text{ T.U. (tritium unit)} = 1 \text{ atom of } ^3\text{H per } 10^{18} \text{ H atoms}$$

$$= 3.23 \times 10^{-3} \text{ pCi/mL}$$

$$= 7.17 \times 10^{-3} \text{ dpm/mL}$$

Decay-correction date is 7 Sept. 1983 (83250).

L: Lysimeter samples. Several types of suction lysimeters, installed at various depths, were sampled and the water was analyzed for tritium. A6, C12, D6, E12, F5, G11, and L-100 are fully buried units with access lines to the surface. L-1 and L-2 are partially buried units that allow full inspection of the lysimeter interior.

ND—Not determined—Within the extensive number of samples collected and analyzed, several could not be finalized due to field or laboratory complications.

LANL CCW: LANL analyses of CCW excerpted from their progress reports for comparison purposes. See subsequent LANL reports for additional data.

NP: Neutron probe.

OUT: Outflow. Ditch-water samples were collected near the outflow pipe for RNM-2s.

RC: Resistance cell. Readings normally were the average of a pair of cells at the same location.

RW: Rainwater. Rainwater was collected in a rain gauge located on the instrumented plot.

SAW: Soil-atmosphere water. Soil-atmosphere (gas) moisture was collected at the instrumented plot using a low-flow, gas-extraction system and silica-gel traps. The extraction systems were installed in place of lysimeters L-3 and L-4 (see Fig. 3A-2) after the lysimeters consistently failed to produce moisture samples.

SEOL: Southeastern side of the lake. Pond grab samples were collected prior to the systematic survey of the pond. See Fig. 3A-1.

SOL: Southern side of the lake. Pond grab samples were collected prior to the survey of the pond. See Fig. 3A-1.

T: Tensiometer.

TPS: Toluene plant samples. Plant samples were collected on 83271 and analyzed for tritium using a toluene water-extraction technique. Samples were processed by DRI and analyzed by LLNL; location numbers tabulated are described above. Replicate field samples processed by LLNL are indicated in the FDPW table.

%VM: Percent of water by volume of soil (approximate).

Table B1: Soil moisture data, Cambic area.

(a) Type: RC

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
83192	13,14-5-11	7750	84072	13,14-5-11	6950
83200	13,14-5-11	6900	84079	13,14-5-11	7100
83206	13,14-5-11	6700	84086	13,14-5-11	7200
83213	13,14-5-11	6700	84093	13,14-5-11	7200
83220	13,14-5-11	6250	84100	13,14-5-11	7250
83227	13,14-5-11	6050	84107	13,14-5-11	7100
83241	13,14-5-11	6000	84052	13,14-5-11	6950
83250	13,14-5-11	6000	84114	13,14-5-11	7350
83255	13,14-5-11	6000	84121	13,14-5-11	7800
83262	13,14-5-11	6100	84128	13,14-5-11	7650
83271	13,14-5-11	6100	84135	13,14-5-11	7300
83283	13,14-5-11	6150	84142	13,14-5-11	7500
83297	13,14-5-11	6150	84151	13,14-5-11	7300
83305	13,14-5-11	6400	84156	13,14-5-11	7250
83311	13,14-5-11	5900	84163	13,14-5-11	7250
83318	13,14-5-11	6100	84170	13,14-5-11	7350
83325	13,14-5-11	5850	84177	13,14-5-11	7250
83332	13,14-5-11	6150	84184	13,14-5-11	7100
83339	13,14-5-11	6250	84191	13,14-5-11	7250
83346	13,14-5-11	6250	84198	13,14-5-11	7400
83353	13,14-5-11	6250	84205	13,14-5-11	7400
83361	13,14-5-11	6200	84212	13,14-5-11	7800
84003	13,14-5-11	6350	84219	13,14-5-11	7400
84009	13,14-5-11	6350	84226	13,14-5-11	6350
84016	13,14-5-11	6350	84241	13,14-5-11	7000
84030	13,14-5-11	6700	84248	13,14-5-11	6600
84037	13,14-5-11	6500	84254	13,14-5-11	6350
84044	13,14-5-11	7000	84261	13,14-5-11	6800
84052	13,14-5-11	6950	84268	13,14-5-11	6800
84058	13,14-5-11	7150	84275	13,14-5-11	7000
84065	13,14-5-11	7150	84282	13,14-5-11	9900

^a Final two numbers of the Sample # identify the location and depth of the instrument. See Sec. 3 and references cited therein for details of locations and descriptions.

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
84290	13,14-5-11	6500	85231	13,14-5-11	6400
84296	13,14-5-11	6500	85238	13,14-5-11	6100
84303	13,14-5-11	6575	85246	13,14-5-11	5550
84310	13,14-5-11	6500	85252	13,14-5-11	5250
84318	13,14-5-11	6500	85259	13,14-5-11	5650
84324	13,14-5-11	6500	85266	13,14-5-11	5450
84331	13,14-5-11	6600	85273	13,14-5-11	5200
84338	13,14-5-11	6500	85287	13,14-5-11	5300
84345	13,14-5-11	6500	85294	13,14-5-11	5400
84352	13,14-5-11	6500	85301	13,14-5-11	4900
84361	13,14-5-11	6500	85308	13,14-5-11	5200
85002	13,14-5-11	7200	85316	13,14-5-11	5000
85009	13,14-5-11	6700	85322	13,14-5-11	5550
85014	13,14-5-11	6850	85329	13,14-5-11	5000
85021	13,14-5-11	6800	85343	13,14-5-11	4900
85029	13,14-5-11	6950	85350	13,14-5-11	3300
85035	13,14-5-11	7250	85357	13,14-5-11	5450
85042	13,14-5-11	7500	85364	13,14-5-11	4850
85056	13,14-5-11	7500	86006	13,14-5-11	5550
85062	13,14-5-11	7500	86013	13,14-5-11	5700
85070	13,14-5-11	7800	86027	13,14-5-11	5950
85077	13,14-5-11	7600	86034	13,14-5-11	5750
85084	13,14-5-11	8200	86041	13,14-5-11	5300
85091	13,14-5-11	8250	86049	13,14-5-11	5900
85098	13,14-5-11	8150	86055	13,14-5-11	6400
85112	13,14-5-11	8050	86062	13,14-5-11	6250
85119	13,14-5-11	8050	86069	13,14-5-11	6250
85126	13,14-5-11	7750	86076	13,14-5-11	5050
85133	13,14-5-11	7750	86083	13,14-5-11	6500
85140	13,14-5-11	7900	86090	13,14-5-11	6500
85148	13,14-5-11	8100	86097	13,14-5-11	7050
85154	13,14-5-11	8550	86104	13,14-5-11	6800
85161	13,14-5-11	8450	86111	13,14-5-11	6800
85168	13,14-5-11	8500	86118	13,14-5-11	6850
85182	13,14-5-11	7950	83192	22-5-5	7750
85189	13,14-5-11	8000	83200	22-5-5	6800
85196	13,14-5-11	7000	83206	22-5-5	6000
85217	13,14-5-11	6250	83213	22-5-5	6000
85224	13,14-5-11	6150	83220	22-5-5	6000

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
83227	22-5-5	5500	84163	22-5-5	2800
83241	22-5-5	3500	84170	22-5-5	2800
83250	22-5-5	3300	84177	22-5-5	2800
83255	22-5-5	3300	84184	22-5-5	2700
83262	22-5-5	3200	84191	22-5-5	2300
83271	22-5-5	3300	84198	22-5-5	2700
83283	22-5-5	3400	84205	22-5-5	2700
83297	22-5-5	3400	84212	22-5-5	2500
83305	22-5-5	3200	84219	22-5-5	2500
83311	22-5-5	3200	84226	22-5-5	2400
83318	22-5-5	3200	84241	22-5-5	2100
83325	22-5-5	3200	84248	22-5-5	2100
83332	22-5-5	3400	84254	22-5-5	2200
83339	22-5-5	3400	84261	22-5-5	2200
83346	22-5-5	3500	84268	22-5-5	2200
83353	22-5-5	3500	84275	22-5-5	2300
83361	22-5-5	3600	84282	22-5-5	2500
84003	22-5-5	3600	84290	22-5-5	2400
84009	22-5-5	3500	84296	22-5-5	3600
84016	22-5-5	3600	84303	22-5-5	2700
84030	22-5-5	3700	84310	22-5-5	2800
84037	22-5-5	3500	84318	22-5-5	2800
84044	22-5-5	3600	84324	22-5-5	2800
84052	22-5-5	3600	84331	22-5-5	3000
84058	22-5-5	3600	84338	22-5-5	3000
84065	22-5-5	3500	84345	22-5-5	3000
84072	22-5-5	3500	84352	22-5-5	3000
84079	22-5-5	3400	84361	22-5-5	3000
84086	22-5-5	3400	85002	22-5-5	3000
84093	22-5-5	3200	85009	22-5-5	3000
84100	22-5-5	3400	85014	22-5-5	3000
84107	22-5-5	3500	85021	22-5-5	3000
84114	22-5-5	3500	85029	22-5-5	3100
84121	22-5-5	3500	85035	22-5-5	3200
84128	22-5-5	3500	85042	22-5-5	3200
84135	22-5-5	3200	85056	22-5-5	3200
84142	22-5-5	3000	85062	22-5-5	3000
84151	22-5-5	3000	85070	22-5-5	3100
84156	22-5-5	2800	85077	22-5-5	3000

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
85084	22-5-5	3100	86041	22-5-5	4200
85091	22-5-5	3000	86049	22-5-5	4500
85098	22-5-5	3100	86055	22-5-5	4500
85112	22-5-5	2900	86062	22-5-5	4200
85119	22-5-5	2900	86069	22-5-5	4000
85126	22-5-5	2800	86076	22-5-5	4000
85133	22-5-5	2800	86083	22-5-5	3800
85140	22-5-5	3000	86090	22-5-5	3600
85148	22-5-5	3400	86097	22-5-5	3500
85154	22-5-5	3100	86104	22-5-5	3500
85161	22-5-5	3400	86111	22-5-5	3500
85168	22-5-5	3500	86118	22-5-5	3400
85182	22-5-5	3300	83192	15,16-11-11	20400
85189	22-5-5	3500	83200	15,16-11-11	11650
85196	22-5-5	3600	83206	15,16-11-11	10400
85217	22-5-5	6900	83213	15,16-11-11	6550
85224	22-5-5	10000	83220	15,16-11-11	6250
85231	22-5-5	20000	83227	15,16-11-11	4800
85238	22-5-5	15000	83241	15,16-11-11	3950
85246	22-5-5	79000	83250	15,16-11-11	3750
85252	22-5-5	5800	83255	15,16-11-11	3700
85259	22-5-5	5500	83262	15,16-11-11	3450
85266	22-5-5	4700	83271	15,16-11-11	3150
85273	22-5-5	4400	83283	15,16-11-11	3100
85287	22-5-5	4100	83297	15,16-11-11	3100
85294	22-5-5	3600	83305	15,16-11-11	3050
85301	22-5-5	3900	83311	15,16-11-11	3000
85308	22-5-5	3900	83318	15,16-11-11	3000
85316	22-5-5	3800	83325	15,16-11-11	2900
85322	22-5-5	4300	83332	15,16-11-11	2900
85329	22-5-5	4200	83339	15,16-11-11	2900
85343	22-5-5	4000	83346	15,16-11-11	2900
85350	22-5-5	4200	83353	15,16-11-11	2900
85357	22-5-5	4400	83361	15,16-11-11	2900
85364	22-5-5	4400	84003	15,16-11-11	2900
86006	22-5-5	4400	84009	15,16-11-11	2900
86013	22-5-5	4400	84016	15,16-11-11	2900
86027	22-5-5	4600	84030	15,16-11-11	3150
86034	22-5-5	4500	84037	15,16-11-11	3000

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
84044	15,16-11-11	3050	84324	15,16-11-11	2450
84052	15,16-11-11	3000	84331	15,16-11-11	2600
84058	15,16-11-11	3050	84338	15,16-11-11	2750
84065	15,16-11-11	3100	84345	15,16-11-11	2650
84072	15,16-11-11	2950	84352	15,16-11-11	2650
84079	15,16-11-11	3050	84361	15,16-11-11	2650
84086	15,16-11-11	3100	85002	15,16-11-11	2900
84093	15,16-11-11	3000	85009	15,16-11-11	2700
84100	15,16-11-11	3300	85014	15,16-11-11	2950
84107	15,16-11-11	3000	85021	15,16-11-11	2950
84114	15,16-11-11	3000	85029	15,16-11-11	2950
84121	15,16-11-11	3000	85035	15,16-11-11	3000
84128	15,16-11-11	3000	85042	15,16-11-11	3100
84135	15,16-11-11	3000	85056	15,16-11-11	3150
84142	15,16-11-11	2950	85062	15,16-11-11	3150
84151	15,16-11-11	2950	85070	15,16-11-11	3150
84156	15,16-11-11	2900	85077	15,16-11-11	3100
84163	15,16-11-11	2800	85084	15,16-11-11	3200
84170	15,16-11-11	2850	85091	15,16-11-11	3150
84177	15,16-11-11	2800	85098	15,16-11-11	3150
84184	15,16-11-11	2600	85112	15,16-11-11	3100
84191	15,16-11-11	2750	85119	15,16-11-11	3100
84198	15,16-11-11	2800	85126	15,16-11-11	3050
84205	15,16-11-11	2800	85133	15,16-11-11	3250
84212	15,16-11-11	2700	85140	15,16-11-11	3050
84219	15,16-11-11	2700	85148	15,16-11-11	3100
84226	15,16-11-11	2650	85154	15,16-11-11	3050
84241	15,16-11-11	2500	85161	15,16-11-11	3100
84248	15,16-11-11	2650	85168	15,16-11-11	3100
84254	15,16-11-11	2550	85182	15,16-11-11	3000
84261	15,16-11-11	2650	85189	15,16-11-11	3000
84268	15,16-11-11	2650	85196	15,16-11-11	3000
84275	15,16-11-11	2600	85217	15,16-11-11	2900
84282	15,16-11-11	2650	85224	15,16-11-11	2950
84290	15,16-11-11	2500	85231	15,16-11-11	2900
84296	15,16-11-11	2600	85238	15,16-11-11	2900
84303	15,16-11-11	2500	85246	15,16-11-11	2950
84310	15,16-11-11	2600	85252	15,16-11-11	2750
84318	15,16-11-11	2600	85259	15,16-11-11	3050

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
85266	15,16-11-11	2800	83271	18,19-11-5	67500
85273	15,16-11-11	2850	83283	18,19-11-5	67500
85287	15,16-11-11	2950	83297	18,19-11-5	65000
85294	15,16-11-11	2700	83305	18,19-11-5	62000
85301	15,16-11-11	2850	83311	18,19-11-5	64500
85308	15,16-11-11	2950	83318	18,19-11-5	62000
85316	15,16-11-11	2800	83325	18,19-11-5	62500
85322	15,16-11-11	3150	83332	18,19-11-5	62500
85329	15,16-11-11	2950	83339	18,19-11-5	62500
85343	15,16-11-11	2950	83346	18,19-11-5	62000
85350	15,16-11-11	3100	83353	18,19-11-5	61500
85357	15,16-11-11	3250	83361	18,19-11-5	60500
85364	15,16-11-11	3100	84003	18,19-11-5	62500
86006	15,16-11-11	3150	84009	18,19-11-5	61500
86013	15,16-11-11	3150	84016	18,19-11-5	59750
86027	15,16-11-11	3400	84030	18,19-11-5	63500
86034	15,16-11-11	3350	84037	18,19-11-5	62500
86041	15,16-11-11	3050	84044	18,19-11-5	62500
86049	15,16-11-11	3550	84052	18,19-11-5	63000
86055	15,16-11-11	3650	84058	18,19-11-5	62500
86062	15,16-11-11	3600	84065	18,19-11-5	62500
86069	15,16-11-11	3500	84072	18,19-11-5	61500
86076	15,16-11-11	3600	84079	18,19-11-5	61500
86083	15,16-11-11	3700	84086	18,19-11-5	60000
86090	15,16-11-11	3650	84093	18,19-11-5	59500
86097	15,16-11-11	3550	84100	18,19-11-5	59500
86104	15,16-11-11	3600	84107	18,19-11-5	58500
86111	15,16-11-11	3700	84114	18,19-11-5	55500
86118	15,16-11-11	3550	84121	18,19-11-5	57500
83192	18,19-11-5	112500	84128	18,19-11-5	56500
83200	18,19-11-5	105000	84135	18,19-11-5	52500
83206	18,19-11-5	95000	84142	18,19-11-5	52000
83213	18,19-11-5	90000	84151	18,19-11-5	52500
83220	18,19-11-5	82500	84156	18,19-11-5	51500
83227	18,19-11-5	110000	84163	18,19-11-5	49500
83241	18,19-11-5	74000	84170	18,19-11-5	51500
83250	18,19-11-5	72500	84177	18,19-11-5	53000
83255	18,19-11-5	72500	84184	18,19-11-5	53000
83262	18,19-11-5	67000	84191	18,19-11-5	48000

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
84198	18,19-11-5	50000	85126	18,19-11-5	2700
84205	18,19-11-5	45000	85133	18,19-11-5	2900
84212	18,19-11-5	43000	85140	18,19-11-5	3000
84219	18,19-11-5	42000	85148	18,19-11-5	1150
84226	18,19-11-5	38500	85154	18,18-11-5	3450
84241	18,19-11-5	32500	85161	18,19-11-5	3650
84248	18,19-11-5	30000	85168	18,19-11-5	4850
84254	18,19-11-5	29000	85182	18,19-11-5	13000
84261	18,19-11-5	25000	85189	18,19-11-5	13000
84268	18,19-11-5	25000	85196	18,19-11-5	12900
84275	18,19-11-5	18000	85217	18,19-11-5	33500
84282	18,19-11-5	18000	85224	18,19-11-5	43500
84290	18,19-11-5	14000	85231	18,19-11-5	49500
84296	18,19-11-5	15000	85238	18,19-11-5	55500
84303	18,19-11-5	14000	85246	18,19-11-5	56500
84310	18,19-11-5	14500	85252	18,19-11-5	55000
84318	18,19-11-5	14000	85259	18,19-11-5	60500
84324	18,19-11-5	14000	85273	18,19-11-5	62000
84331	18,19-11-5	14000	85287	18,19-11-5	60000
84338	18,19-11-5	15750	85294	18,19-11-5	38000
84345	18,19-11-5	15750	85301	18,19-11-5	41000
84352	18,19-11-5	16000	85308	18,19-11-5	42000
84361	18,19-11-5	17500	85316	18,19-11-5	41000
85002	18,19-11-5	17500	85322	18,19-11-5	41000
85009	18,19-11-5	5850	85329	18,19-11-5	38000
85014	18,19-11-5	4700	85343	18,19-11-5	59500
85021	18,19-11-5	3650	85350	18,19-11-5	59500
85029	18,19-11-5	3350	85357	18,19-11-5	60000
85035	18,19-11-5	2950	85364	18,19-11-5	56000
85042	18,19-11-5	2800	86006	18,19-11-5	56500
85056	18,19-11-5	2600	86013	18,19-11-5	51500
85062	18,19-11-5	2500	86027	18,19-11-5	54000
85070	18,19-11-5	2500	86034	18,19-11-5	53500
85077	18,19-11-5	2500	86041	18,19-11-5	53500
85084	18,19-11-5	2500	86049	18,19-11-5	49500
85091	18,19-11-5	2400	86055	18,19-11-5	49500
85098	18,19-11-5	2450	86062	18,19-11-5	39250
85112	18,19-11-5	2600	86069	18,19-11-5	46000
85119	18,19-11-5	2700	86076	18,19-11-5	44500

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
86083	18,19-11-5	44500	84086	9,12-11-7	4850
86090	18,19-11-5	44000	84093	9,12-11-7	4900
86097	18,19-11-5	43000	84100	9,12-11-7	4950
86104	18,19-11-5	40000	84107	9,12-11-7	5050
86111	18,19-11-5	41000	84114	9,12-11-7	4900
86118	18,19-11-5	38000	84121	9,12-11-7	5100
83192	9,12-11-7	4500	84128	9,12-11-7	5150
83200	9,12-11-7	4100	84135	9,12-11-7	5150
83206	9,12-11-7	3700	84142	9,12-11-7	4850
83213	9,12-11-7	3550	84151	9,12-11-7	4950
83220	9,12-11-7	3500	84156	9,12-11-7	4800
83227	9,12-11-7	3400	84163	9,12-11-7	4750
83241	9,12-11-7	3300	84170	9,12-11-7	4900
83250	9,12-11-7	3200	84177	9,12-11-7	4750
83255	9,12-11-7	3200	84184	9,12-11-7	4750
83262	9,12-11-7	3150	84191	9,12-11-7	4750
83271	9,12-11-7	3150	84198	9,12-11-7	4750
83283	9,12-11-7	3150	84205	9,12-11-7	4750
83297	9,12-11-7	3450	84212	9,12-11-7	4750
83305	9,12-11-7	3450	84219	9,12-11-7	4850
83311	9,12-11-7	3450	84226	9,12-11-7	4800
83318	9,12-11-7	3600	84241	9,12-11-7	4800
83325	9,12-11-7	3600	84248	9,12-11-7	4850
83332	9,12-11-7	3700	84254	9,12-11-7	4800
83339	9,12-11-7	3800	84261	9,12-11-7	4800
83346	9,12-11-7	3950	84268	9,12-11-7	4800
83353	9,12-11-7	3950	84275	9,12-11-7	4900
83361	9,12-11-7	4200	84282	9,12-11-7	5050
84003	9,12-11-7	4400	84290	9,12-11-7	5250
84009	9,12-11-7	4350	84296	9,12-11-7	5250
84016	9,12-11-7	4400	84303	9,12-11-7	5450
84030	9,12-11-7	4750	84310	9,12-11-7	5650
84037	9,12-11-7	5000	84318	9,12-11-7	5750
84044	9,12-11-7	4800	84324	9,12-11-7	5750
84052	9,12-11-7	4850	84331	9,12-11-7	5750
84058	9,12-11-7	5100	84338	9,12-11-7	6200
84065	9,12-11-7	4850	84345	9,12-11-7	6500
84072	9,12-11-7	4850	84352	9,12-11-7	6400
84079	9,12-11-7	4900	84361	9,12-11-7	6500

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
85002	9,12-11-7	6750	85316	9,12-11-7	2900
85009	9,12-11-7	7150	85322	9,12-11-7	3150
85014	9,12-11-7	7150	85329	9,12-11-7	3100
85021	9,12-11-7	7150	85343	9,12-11-7	3250
85029	9,12-11-7	7500	85350	9,12-11-7	3400
85035	9,12-11-7	7150	85357	9,12-11-7	3450
85042	9,12-11-7	7300	85364	9,12-11-7	3550
85056	9,12-11-7	7350	86006	9,12-11-7	3550
85062	9,12-11-7	7500	86013	9,12-11-7	3550
85070	9,12-11-7	7350	86027	9,12-11-7	3850
85077	9,12-11-7	7000	86034	9,12-11-7	3800
85084	9,12-11-7	7450	86041	9,12-11-7	3450
85091	9,12-11-7	7300	86049	9,12-11-7	3950
85098	9,12-11-7	6850	86055	9,12-11-7	4100
85112	9,12-11-7	6600	86062	9,12-11-7	4300
85119	9,12-11-7	6550	86069	9,12-11-7	3800
85126	9,12-11-7	6450	86076	9,12-11-7	3950
85133	9,12-11-7	6250	86083	9,12-11-7	4000
85140	9,12-11-7	6200	86090	9,12-11-7	3850
85148	9,12-11-7	5900	86097	9,12-11-7	4050
85154	9,12-11-7	5900	86104	9,12-11-7	3800
85161	9,12-11-7	5700	86111	9,12-11-7	3850
85168	9,12-11-7	7050	86118	9,12-11-7	3950
85182	9,12-11-7	4550	83192	24,25-11-3	1150000
85189	9,12-11-7	5200	83200	24,25-11-3	1200000
85196	9,12-11-7	16500	83206	24,25-11-3	1125000
85217	9,12-11-7	22500	83213	24,25-11-3	1125000
85224	9,12-11-7	2350	83220	24,25-11-3	825000
85231	9,12-11-7	2600	83227	24,25-11-3	80000
85238	9,12-11-7	2550	83241	24,25-11-3	800000
85246	9,12-11-7	2600	83250	24,25-11-3	550000
85252	9,12-11-7	2100	83255	24,25-11-3	450000
85259	9,12-11-7	2800	83262	24,25-11-3	240000
85266	9,12-11-7	2600	83271	24,25-11-3	112500
85273	9,12-11-7	2650	83283	24,25-11-3	107000
85287	9,12-11-7	2750	83297	24,25-11-3	115000
85294	9,12-11-7	2600	83305	24,25-11-3	107500
85301	9,12-11-7	2900	83311	24,25-11-3	110000
85308	9,12-11-7	2950	83318	24,25-11-3	122500

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
83325	24,25-11-3	125000	84248	24,25-11-3	90000
83332	24,25-11-3	140000	84254	24,25-11-3	72500
83339	24,25-11-3	145000	84261	24,25-11-3	60000
83346	24,25-11-3	160000	84268	24,25-11-3	60000
83353	24,25-11-3	145000	84275	24,25-11-3	60000
83361	24,25-11-3	155000	84282	24,25-11-3	64000
84003	24,25-11-3	155000	84290	24,25-11-3	67500
84009	24,25-11-3	145000	84296	24,25-11-3	66500
84016	24,25-11-3	147500	84303	24,25-11-3	72500
84030	24,25-11-3	145000	84310	24,25-11-3	80000
84037	24,25-11-3	140000	84318	24,25-11-3	80000
84044	24,25-11-3	130000	84324	24,25-11-3	82500
84052	24,25-11-3	135000	84331	24,25-11-3	85500
84058	24,25-11-3	130000	84338	24,25-11-38	8000
84065	24,25-11-3	125000	84345	24,25-11-3	95000
84072	24,25-11-3	125000	84352	24,25-11-3	95000
84079	24,25-11-3	112500	84361	24,25-11-3	45500
84086	24,25-11-3	110000	85002	24,25-11-3	4000
84093	24,25-11-3	109000	85009	24,25-11-3	2150
84100	24,25-11-3	104000	85014	24,25-11-3	1900
84107	24,25-11-3	94000	85021	24,25-11-3	1900
84114	24,25-11-3	95000	85029	24,25-11-3	1850
84121	24,25-11-3	95000	85035	24,25-11-3	2050
84128	24,25-11-3	94000	85042	24,25-11-3	2300
84135	24,25-11-3	86500	85056	24,25-11-3	2100
84142	24,25-11-3	86000	85062	24,25-11-3	2000
84151	24,25-11-3	83500	85070	24,25-11-3	2200
84156	24,25-11-3	89000	85077	24,25-11-3	2050
84163	24,25-11-3	94000	85084	24,25-11-3	2050
84170	24,25-11-3	102500	85091	24,25-11-3	2200
84177	24,25-11-3	110000	85098	24,25-11-3	2050
84184	24,25-11-3	115000	85112	24,25-11-3	2050
84191	24,25-11-3	120000	85119	24,25-11-3	2150
84198	24,25-11-3	117500	85126	24,25-11-3	2200
84205	24,25-11-3	117500	85133	24,25-11-3	2650
84212	24,25-11-3	110000	85140	24,25-11-3	4000
84219	24,25-11-3	105000	85148	24,25-11-3	10500
84226	24,25-11-3	96000	85154	24,25-11-3	27000
84241	24,25-11-3	95500	85161	24,25-11-3	67500

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
85168	24,25-11-3	95000	86118	24,25-11-3	300000
85182	24,25-11-3	117500	83192	17,20-21-4	1400000
85189	24,25-11-3	135000	83200	17,20-21-4	1600000
85196	24,25-11-3	150000	83206	17,20-21-4	1750000
85217	24,25-11-3	170000	83213	17,20-21-4	1750000
85224	24,25-11-3	210000	83220	17,20-21-4	750000
85231	24,25-11-3	305000	83227	17,20-21-4	750000
85238	24,25-11-3	325000	83241	17,20-21-4	1500000
85246	24,25-11-3	335000	83250	17,20-21-4	1750000
85252	24,25-11-3	330000	83255	17,20-21-4	1500000
85259	24,25-11-3	385000	83262	17,20-21-4	1250000
85266	24,25-11-3	415000	83271	17,20-21-4	1250000
85273	24,25-11-3	425000	83283	17,20-21-4	1500000
85287	24,25-11-3	490000	83297	17,20-21-4	1500000
85294	24,25-11-3	520000	83305	17,20-21-4	1250000
85301	24,25-11-3	500000	83311	17,20-21-4	1250000
85308	24,25-11-3	560000	83318	17,20-21-4	1250000
85316	24,25-11-3	560000	83325	17,20-21-4	900000
85322	24,25-11-3	430000	83332	17,20-21-4	875000
85329	24,25-11-3	660000	83339	17,20-21-4	900000
85343	24,25-11-3	650000	83346	17,20-21-4	900000
85350	24,25-11-3	650000	83353	17,20-21-4	875000
85357	24,25-11-3	650000	83361	17,20-21-4	1000000
85364	24,25-11-3	650000	84003	17,20-21-4	875000
86006	24,25-11-3	650000	84009	17,20-21-4	1125000
86013	24,25-11-3	650000	84016	17,20-21-4	875000
86027	24,25-11-3	625000	84030	17,20-21-4	1250000
86034	24,25-11-3	600000	84037	17,20-21-4	1000000
86041	24,25-11-3	600000	84044	17,20-21-4	1200000
86049	24,25-11-3	560000	84052	17,20-21-4	1050000
86055	24,25-11-3	550000	84058	17,20-21-4	1200000
86062	24,25-11-3	500000	84065	17,20-21-4	875000
86069	24,25-11-3	425000	84072	17,20-21-4	875000
86076	24,25-11-3	425000	84079	17,20-21-4	875000
86083	24,25-11-3	475000	84086	17,20-21-4	875000
86090	24,25-11-3	385000	84093	17,20-21-4	875000
86097	24,25-11-3	405000	84100	17,20-21-4	875000
86104	24,25-11-3	345000	84107	17,20-21-4	875000
86111	24,25-11-3	340000	84114	17,20-21-4	800000

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
84121	17,20-21-4	875000	85035	17,20-21-4	12350
84128	17,20-21-4	875000	85042	17,20-21-4	10800
84135	17,20-21-4	875000	85056	17,20-21-4	10500
84142	17,20-21-4	875000	85062	17,20-21-4	8900
84151	17,20-21-4	900000	85070	17,20-21-4	8900
84156	17,20-21-4	875000	85077	17,20-21-4	9300
84163	17,20-21-4	900000	85084	17,20-21-4	8300
84170	17,20-21-4	1000000	85091	17,20-21-4	7860
84177	17,20-21-4	900000	85098	17,20-21-4	7450
84184	17,20-21-4	1250000	85112	17,20-21-4	9950
84191	17,20-21-4	1150000	85119	17,20-21-4	14750
84198	17,20-21-4	1150000	85126	17,20-21-4	54000
84205	17,20-21-4	1150000	85133	17,20-21-4	101000
84212	17,20-21-4	1150000	85140	17,20-21-4	180000
84219	17,20-21-4	1000000	85148	17,20-21-4	265000
84226	17,20-21-4	1150000	85154	17,20-21-4	285000
84241	17,20-21-4	1000000	85161	17,20-21-4	335000
84248	17,20-21-4	900000	85168	17,20-21-4	385000
84254	17,20-21-4	875000	85182	17,20-21-4	425000
84261	17,20-21-4	875000	85189	17,20-21-4	425000
84268	17,20-21-4	875000	85196	17,20-21-4	535000
84275	17,20-21-4	750000	85217	17,20-21-4	110000
84282	17,20-21-4	750000	85224	17,20-21-4	500000
84290	17,20-21-4	800000	85231	17,20-21-4	1450000
84296	17,20-21-4	800000	85238	17,20-21-4	1550000
84303	17,20-21-4	800000	85246	17,20-21-4	1500000
84310	17,20-21-4	800000	85252	17,20-21-4	1500000
84318	17,20-21-4	875000	85259	17,20-21-4	1550000
84324	17,20-21-4	850000	85266	17,20-21-4	1600000
84331	17,20-21-4	1150000	85273	17,20-21-4	1525000
84338	17,20-21-4	1150000	85287	17,20-21-4	1550000
84345	17,20-21-4	1125000	85294	17,20-21-4	1250000
84352	17,20-21-4	1150000	85301	17,20-21-4	1100000
84361	17,20-21-4	1500000	85308	17,20-21-4	2000000
85002	17,20-21-4	1200000	85316	17,20-21-4	1550000
85009	17,20-21-4	136500	85322	17,20-21-4	2200000
85014	17,20-21-4	26500	85329	17,20-21-4	2150000
85021	17,20-21-4	16500	85343	17,20-21-4	2000000
85029	17,20-21-4	12500	85350	17,20-21-4	2250000

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
85357	17,20-21-4	1900000	83361	30,31-21-10	89500
85364	17,20-21-4	2125000	84003	30,31-21-10	89500
86006	17,20-21-4	2125000	84009	30,31-21-10	90000
86013	17,20-21-4	2150000	84016	30,31-21-10	90000
86027	17,20-21-4	2400000	84030	30,31-21-10	92500
86034	17,20-21-4	2125000	84037	30,31-21-10	90000
86041	17,20-21-4	1600000	84044	30,31-21-10	90000
86049	17,20-21-4	2175000	84052	30,31-21-10	95000
86055	17,20-21-4	2000000	84058	30,31-21-10	95000
86062	17,20-21-4	1600000	84065	30,31-21-10	95000
86069	17,20-21-4	2000000	84072	30,31-21-10	87500
86076	17,20-21-4	1500000	84079	30,31-21-10	90000
86083	17,20-21-4	1500000	84086	30,31-21-10	90000
86090	17,20-21-4	2000000	84093	30,31-21-10	90000
86097	17,20-21-4	1650000	84100	30,31-21-10	92500
86104	17,20-21-4	1500000	84107	30,31-21-10	90000
86111	17,20-21-4	1500000	84114	30,31-21-10	90000
86118	17,20-21-4	1500000	84121	30,31-21-10	89000
83192	30,31-21-10	160000	84128	30,31-21-10	86500
83200	30,31-21-10	155000	84135	30,31-21-10	85000
83206	30,31-21-10	140000	84142	30,31-21-10	84000
83213	30,31-21-10	135000	84151	30,31-21-10	84000
83220	30,31-21-10	135000	84156	30,31-21-10	83500
83227	30,31-21-10	120000	84163	30,31-21-10	80000
83241	30,31-21-10	115000	84170	30,31-21-10	82500
83250	30,31-21-10	110000	84177	30,31-21-10	75000
83255	30,31-21-10	110000	84184	30,31-21-10	80000
83262	30,31-21-10	102500	84191	30,31-21-10	74000
83271	30,31-21-10	100000	84198	30,31-21-10	75000
83283	30,31-21-10	97500	84205	30,31-21-10	74000
83297	30,31-21-10	92500	84212	30,31-21-10	70000
83305	30,31-21-10	90000	84219	30,31-21-10	68500
83311	30,31-21-10	90000	84226	30,31-21-10	66500
83318	30,31-21-10	90000	84241	30,31-21-10	65000
83325	30,31-21-10	90000	84248	30,31-21-10	64000
83332	30,31-21-10	87500	84254	30,31-21-10	64000
83339	30,31-21-10	87500	84261	30,31-21-10	60000
83346	30,31-21-10	90000	84268	30,31-21-10	60000
83353	30,31-21-10	90000	84275	30,31-21-10	57500

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
84282	30,31-21-10	600000	85224	30,31-21-10	78000
84290	30,31-21-10	56500	85231	30,31-21-10	98000
84296	30,31-21-10	56500	85238	30,31-21-10	73000
84303	30,31-21-10	56500	85246	30,31-21-10	51500
84310	30,31-21-10	55000	85252	30,31-21-10	34000
84318	30,31-21-10	57000	85259	30,31-21-10	30000
84324	30,31-21-10	57000	85266	30,31-21-10	28000
84331	30,31-21-10	55000	85273	30,31-21-10	27500
84338	30,31-21-10	56500	85287	30,31-21-10	27500
84345	30,31-21-10	56000	85294	30,31-21-10	24500
84352	30,31-21-10	57000	85301	30,31-21-10	27500
84361	30,31-21-10	58500	85308	30,31-21-10	27500
85002	30,31-21-10	58500	85316	30,31-21-10	26500
85009	30,31-21-10	58500	85322	30,31-21-10	28500
85014	30,31-21-10	60500	85329	30,31-21-10	27500
85021	30,31-21-10	60500	85343	30,31-21-10	28000
85029	30,31-21-10	63500	85350	30,31-21-10	27500
85035	30,31-21-10	62000	85357	30,31-21-10	28500
85042	30,31-21-10	62500	85364	30,31-21-10	28500
85056	30,31-21-10	62500	86006	30,31-21-10	28500
85062	30,31-21-10	70000	86013	30,31-21-10	29500
85070	30,31-21-10	65000	86027	30,31-21-10	31000
85077	30,31-21-10	63500	86034	30,31-21-10	31000
85084	30,31-21-10	65500	86041	30,31-21-10	28500
85091	30,31-21-10	63500	86049	30,31-21-10	31000
85098	30,31-21-10	63500	86055	30,31-21-10	32500
85112	30,31-21-10	63500	86062	30,31-21-10	32000
85119	30,31-21-10	62000	86069	30,31-21-10	31500
85126	30,31-21-10	61000	86076	30,31-21-10	32500
85133	30,31-21-10	60000	86083	30,31-21-10	33000
85140	30,31-21-10	63500	86090	30,31-21-10	32500
85148	30,31-21-10	57500	86097	30,31-21-10	32500
85154	30,31-21-10	55000	86104	30,31-21-10	33000
85161	30,31-21-10	53500	86111	30,31-21-10	32500
85168	30,31-21-10	51500	86118	30,31-21-10	33500
85182	30,31-21-10	47500	98192	1,2-11-15-	660
85189	30,31-21-10	45000	83200	1,2-11-15	1010
85196	30,31-21-10	44000	83206	1,2-11-15	1250
85217	30,31-21-10	62000	83213	1,2-11-15	1450

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
83220	1,2-11-15	1550	84156	1,2-11-15	2450
83227	1,2-11-15	1700	84163	1,2-11-15	2450
83241	1,2-11-15	1725	84170	1,2-11-15	2350
83250	1,2-11-15	1740	84177	1,2-11-15	2450
83255	1,2-11-15	1790	84184	1,2-11-15	2400
83262	1,2-11-15	1800	84191	1,2-11-15	2450
83271	1,2-11-15	1800	84198	1,2-11-15	2550
83283	1,2-11-15	1900	84205	1,2-11-15	3000
83297	1,2-11-15	1900	84212	1,2-11-15	2450
83305	1,2-11-15	1950	84219	1,2-11-15	2450
83311	1,2-11-15	1850	84226	1,2-11-15	2450
83318	1,2-11-15	1900	84241	1,2-11-15	2450
83325	1,2-11-15	1400	84248	1,2-11-15	2500
83332	1,2-11-15	1850	84254	1,2-11-15	2500
83339	1,2-11-15	1900	84261	1,2-11-15	2450
83346	1,2-11-15	1950	84268	1,2-11-15	2450
83353	1,2-11-15	1950	84275	1,2-11-15	2500
83361	1,2-11-15	1875	84282	1,2-11-15	2500
84003	1,2-11-15	1875	84290	1,2-11-15	2500
84009	1,2-11-15	1925	84296	1,2-11-15	2350
84016	1,2-11-15	1925	84303	1,2-11-15	2550
84030	1,2-11-15	2100	84310	1,2-11-15	2600
84037	1,2-11-15	2050	84318	1,2-11-15	2500
84044	1,2-11-15	2013	84324	1,2-11-15	2550
84052	1,2-11-15	2200	84331	1,2-11-15	2450
84058	1,2-11-15	2200	84338	1,2-11-15	2600
84065	1,2-11-15	2400	84345	1,2-11-15	2600
84072	1,2-11-15	2350	84352	1,2-11-15	2500
84079	1,2-11-15	2300	84361	1,2-11-15	2600
84086	1,2-11-15	2300	85002	1,2-11-15	2650
84093	1,2-11-15	2300	85009	1,2-11-15	2700
84100	1,2-11-15	2350	85014	1,2-11-15	2750
84107	1,2-11-15	2450	85021	1,2-11-15	2800
84114	1,2-11-15	2450	85029	1,2-11-15	2850
84121	1,2-11-15	2450	85035	1,2-11-15	2750
84128	1,2-11-15	2350	85042	1,2-11-15	2950
84135	1,2-11-15	2450	85056	1,2-11-15	3000
84142	1,2-11-15	2450	85062	1,2-11-15	3050
84151	1,2-11-15	2450	85070	1,2-11-15	3150

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
85077	1,2-11-15	3900	86034	1,2-11-15	3200
85084	1,2-11-15	3150	86041	1,2-11-15	3150
85091	1,2-11-15	3250	86049	1,2-11-15	3700
85098	1,2-11-15	3250	86055	1,2-11-15	3300
85112	1,2-11-15	3250	86062	1,2-11-15	3300
85119	1,2-11-15	3250	86069	1,2-11-15	3200
85126	1,2-11-15	3200	86076	1,2-11-15	3250
85133	1,2-11-15	3300	86083	1,2-11-15	3400
85140	1,2-11-15	3300	86090	1,2-11-15	3300
85148	1,2-11-15	3250	86097	1,2-11-15	3350
85154	1,2-11-15	3200	86104	1,2-11-15	3350
85161	1,2-11-15	3250	86111	1,2-11-15	3600
85168	1,2-11-15	3200	86118	1,2-11-15	4000
85182	1,2-11-15	3250	83192	7,8-11-10	9000
85189	1,2-11-15	2350	83200	7,8-11-10	8000
85196	1,2-11-15	3200	83206	7,8-11-10	7500
85217	1,2-11-15	2950	83213	7,8-11-10	7000
85224	1,2-11-15	2950	83220	7,8-11-10	6050
85231	1,2-11-15	2950	83227	7,8-11-10	5900
85238	1,2-11-15	2950	83241	7,8-11-10	5250
85246	1,2-11-15	2950	83250	7,8-11-10	5100
85252	1,2-11-15	2900	83255	7,8-11-10	5000
85259	1,2-11-15	3050	83262	7,8-11-10	6000
85266	1,2-11-15	2950	83271	7,8-11-10	5000
85273	1,2-11-15	2950	83283	7,8-11-10	4650
85287	1,2-11-15	2900	83297	7,8-11-10	4650
85294	1,2-11-15	2900	83305	7,8-11-10	4650
85301	1,2-11-15	3050	83311	7,8-11-10	4650
85308	1,2-11-15	2850	83318	7,8-11-10	4650
85316	1,2-11-15	2800	83325	7,8-11-10	4450
85322	1,2-11-15	2950	83332	7,8-11-10	4300
85329	1,2-11-15	2850	83339	7,8-11-10	4300
85343	1,2-11-15	2800	83346	7,8-11-10	4400
85350	1,2-11-15	2800	83353	7,8-11-10	4300
85357	1,2-11-15	2950	83361	7,8-11-10	4450
85364	1,2-11-15	3000	84003	7,8-11-10	4450
86006	1,2-11-15	3000	84009	7,8-11-10	4500
86013	1,2-11-15	3100	84016	7,8-11-10	4500
86027	1,2-11-15	3200	84030	7,8-11-10	4550

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
84037	7,8-11-10	4550	84318	7,8-11-10	4050
84044	7,8-11-10	4925	84324	7,8-11-10	4050
84052	7,8-11-10	4800	84331	7,8-11-10	4100
84058	7,8-11-10	4850	84338	7,8-11-10	4200
84065	7,8-11-10	4900	84345	7,8-11-10	4350
84072	7,8-11-10	4850	84352	7,8-11-10	4150
84079	7,8-11-10	4900	84361	7,8-11-10	4250
84086	7,8-11-10	4850	85002	7,8-11-10	4100
84093	7,8-11-10	4700	85009	7,8-11-10	4400
84100	7,8-11-10	5150	85014	7,8-11-10	4450
84107	7,8-11-10	4900	85021	7,8-11-10	4500
84114	7,8-11-10	5100	85029	7,8-11-10	4500
84121	7,8-11-10	5300	85035	7,8-11-10	4500
84128	7,8-11-10	5000	85042	7,8-11-10	4550
84135	7,8-11-10	4800	85056	7,8-11-10	4750
84142	7,8-11-10	4900	85062	7,8-11-10	4700
84151	7,8-11-10	4800	85070	7,8-11-10	4950
84156	7,8-11-10	4650	85077	7,8-11-10	4550
84163	7,8-11-10	4650	85084	7,8-11-10	4750
84170	7,8-11-10	4550	85091	7,8-11-10	4650
84177	7,8-11-10	4550	85098	7,8-11-10	4850
84184	7,8-11-10	4500	85112	7,8-11-10	4750
84191	7,8-11-10	4750	85119	7,8-11-10	4600
84198	7,8-11-10	4400	85126	7,8-11-10	4550
84205	7,8-11-10	4400	85133	7,8-11-10	4300
84212	7,8-11-10	4250	85140	7,8-11-10	4500
84219	7,8-11-10	4250	85148	7,8-11-10	4250
84226	7,8-11-10	4050	85154	7,8-11-10	4100
84241	7,8-11-10	4050	85161	7,8-11-10	4150
84248	7,8-11-10	4750	85168	7,8-11-10	4200
84254	7,8-11-10	4050	85182	7,8-11-10	4050
84261	7,8-11-10	4000	85189	7,8-11-10	3750
84268	7,8-11-10	4000	85196	7,8-11-10	3900
84275	7,8-11-10	4150	85217	7,8-11-10	3700
84282	7,8-11-10	4150	85224	7,8-11-10	3700
84290	7,8-11-10	4050	85231	7,8-11-10	3600
84296	7,8-11-10	4000	85238	7,8-11-10	3700
84303	7,8-11-10	4050	85246	7,8-11-10	3600
84310	7,8-11-10	4050	85252	7,8-11-10	3150

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
85259	7,8-11-10	3700	83262	10,11-11-5	120000
85266	7,8-11-10	3650	83271	10,11-11-5	115000
85273	7,8-11-10	3600	83283	10,11-11-5	110000
85287	7,8-11-10	3500	83297	10,11-11-5	105000
85294	7,8-11-10	3800	83305	10,11-11-5	100000
85301	7,8-11-10	3600	83311	10,11-11-5	100000
85308	7,8-11-10	3450	83318	10,11-11-5	100000
85316	7,8-11-10	3350	83325	10,11-11-5	100000
85322	7,8-11-10	3700	83332	10,11-11-5	100000
85329	7,8-11-10	3550	83339	10,11-11-5	100000
85343	7,8-11-10	3500	83346	10,11-11-5	100000
85350	7,8-11-10	3550	83353	10,11-11-5	97500
85357	7,8-11-10	3800	83361	10,11-11-5	100000
85364	7,8-11-10	3650	84003	10,11-11-5	99500
86006	7,8-11-10	3600	84009	10,11-11-5	105000
86013	7,8-11-10	3800	84016	10,11-11-5	97500
86027	7,8-11-10	4150	84030	10,11-11-5	104500
86034	7,8-11-10	3850	84037	10,11-11-5	101500
86041	7,8-11-10	3800	84044	10,11-11-5	100000
86049	7,8-11-10	4000	84052	10,11-11-5	100000
86055	7,8-11-10	4100	84058	10,11-11-5	100000
86062	7,8-11-10	4000	84065	10,11-11-5	100000
86069	7,8-11-10	3900	84072	10,11-11-5	89000
86076	7,8-11-10	4000	84079	10,11-11-5	89000
86083	7,8-11-10	4150	84086	10,11-11-5	88500
86090	7,8-11-10	4050	84093	10,11-11-5	83500
86097	7,8-11-10	4250	84100	10,11-11-5	83000
86104	7,8-11-10	4150	84107	10,11-11-5	78000
86111	7,8-11-10	4250	84114	10,11-11-5	75000
86118	7,8-11-10	4200	84121	10,11-11-5	73500
83192	10,11-11-5	230000	84128	10,11-11-5	68500
83200	10,11-11-5	235000	84135	10,11-11-5	66000
83206	10,11-11-5	250000	84142	10,11-11-5	62500
83213	10,11-11-5	235000	84151	10,11-11-5	58000
83220	10,11-11-5	18500	84156	10,11-11-5	56500
83227	10,11-11-5	17000	84163	10,11-11-5	54500
83241	10,11-11-5	145000	84170	10,11-11-5	51000
83250	10,11-11-5	140000	84177	10,11-11-5	47500
83255	10,11-11-5	130000	84184	10,11-11-5	45000

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
84191	10,11-11-5	42500	85119	10,11-11-5	4050
84198	10,11-11-5	37500	85126	10,11-11-5	3950
84205	10,11-11-5	37500	85133	10,11-11-5	3800
84212	10,11-11-5	36000	85140	10,11-11-5	3800
84219	10,11-11-5	36000	85148	10,11-11-5	3650
84226	10,11-11-5	33000	85154	10,11-11-5	3600
84241	10,11-11-5	33500	85161	10,11-11-5	3550
84248	10,11-11-5	33000	85168	10,11-11-5	3550
84254	10,11-11-5	32000	85182	10,11-11-5	3350
84261	10,11-11-5	30000	85189	10,11-11-5	3450
84268	10,11-11-5	30000	85196	10,11-11-5	3300
84275	10,11-11-5	27000	85217	10,11-11-5	3300
84282	10,11-11-5	25500	85224	10,11-11-5	3500
84290	10,11-11-5	26500	85231	10,11-11-5	3600
84296	10,11-11-5	25500	85238	10,11-11-5	3700
84303	10,11-11-5	24000	85246	10,11-11-5	3850
84310	10,11-11-5	22000	85252	10,11-11-5	4150
84318	10,11-11-5	26000	85259	10,11-11-5	4350
84324	10,11-11-5	24000	85266	10,11-11-5	4400
84331	10,11-11-5	24500	85273	10,11-11-5	4750
84338	10,11-11-5	25500	85287	10,11-11-5	5050
84345	10,11-11-5	24500	85294	10,11-11-5	5900
84352	10,11-11-5	24500	85301	10,11-11-5	5350
84361	10,11-11-5	24500	85308	10,11-11-5	5600
85002	10,11-11-5	25500	85316	10,11-11-5	5600
85009	10,11-11-5	28000	85322	10,11-11-5	6600
85014	10,11-11-5	27000	85329	10,11-11-5	6250
85021	10,11-11-5	21500	85343	10,11-11-5	6550
85029	10,11-11-5	11500	85350	10,11-11-5	6750
85035	10,11-11-5	8000	85357	10,11-11-5	7250
85042	10,11-11-5	7000	85364	10,11-11-5	7100
85056	10,11-11-5	6150	86006	10,11-11-5	7200
85062	10,11-11-5	5550	86013	10,11-11-5	7500
85070	10,11-11-5	5450	86027	10,11-11-5	8000
85077	10,11-11-5	5050	86034	10,11-11-5	7450
85084	10,11-11-5	4850	86041	10,11-11-5	6750
85091	10,11-11-5	4750	86049	10,11-11-5	7750
85098	10,11-11-5	4650	86055	10,11-11-5	7950
85112	10,11-11-5	4150	86062	10,11-11-5	7900

Table B1, Type: RC, continued.

Julian date	Sample# ^a	Reading (Ω)
86069	10,11-11-5	6800
86076	10,11-11-5	7000
86083	10,11-11-5	7350
86090	10,11-11-5	7000

Julian date	Sample# ^a	Reading (Ω)
86097	10,11-11-5	7000
86104	10,11-11-5	6950
86111	10,11-11-5	6850
86118	10,11-11-5	6200

Table B1, continued.

(b) Type: NP

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
83192	11-5	12.00	84086	11-5	—
83200	11-5	12.7	84093	11-5	—
83206	11-5	12.7	84100	11-5	12.8
83213	11-5	12.3	84107	11-5	12.1
83220	11-5	13.0	84114	11-5	12.3
83227	11-5	12.5	84121	11-5	13.3
83241	11-5	13.3	84128	11-5	12.8
83250	11-5	13.0	84135	11-5	13.2
83255	11-5	13.1	84142	11-5	13.5
83262	11-5	15.2	84151	11-5	—
83271	11-5	13.0	84156	11-5	—
83283	11-5	13.7	84163	11-5	—
83297	11-5	13.0	84170	11-5	—
83305	11-5	14.0	84177	11-5	—
83311	11-5	12.9	84184	11-5	—
83318	11-5	13.6	84191	11-5	—
83325	11-5	13.1	84198	11-5	13.6
83332	11-5	13.1	84205	11-5	13.8
83339	11-5	13.1	84212	11-5	—
83346	11-5	13.2	84219	11-5	12.9
83353	11-5	12.7	84226	11-5	13.0
83361	11-5	13.0	83192	11-7.5	15.2
84003	11-5	12.5	83200	11-7.5	15.5
84009	11-5	13.2	83206	11-7.5	15.1
84016	11-5	13.5	83213	11-7.5	14.7
84030	11-5	—	83220	11-7.5	15.5
84037	11-5	—	83227	11-7.5	14.7
84044	11-5	—	83241	11-7.5	14.8
84052	11-5	—	83250	11-7.5	14.9
84058	11-5	—	83255	11-7.5	15.3
84065	11-5	—	83262	11-7.5	13.0
84072	11-5	—	83271	11-7.5	14.9
84079	11-5	—	83283	11-7.5	14.3

^a Final two numbers of the Sample # identify the location and depth of the instrument. See Sec. 3 and references cited therein for details of locations and descriptions.

Table B1, Type: NP, continued.

Julian date	Sample# ^a	Reading (%VM)	Julian date	Sample# ^a	Reading (%VM)
83297	11-7.5	14.4	84212	11-7.5	—
83305	11-7.5	14.6	84219	11-7.5	16.3
83311	11-7.5	14.3	84226	11-7.5	13.3
83318	11-7.5	14.4	83192	11-10	14.8
83325	11-7.5	14.5	83200	11-10	15.0
83332	11-7.5	14.6	83206	11-10	14.8
83339	11-7.5	14.3	83213	11-10	14.6
83346	11-7.5	15.0	83220	11-10	15.3
83353	11-7.5	14.2	83227	11-10	14.5
83361	11-7.5	14.2	83241	11-10	14.7
84003	11-7.5	14.6	83250	11-10	14.7
84009	11-7.5	14.8	83255	11-10	13.3
84016	11-7.5	14.8	83262	11-10	15.1
84030	11-7.5	—	83271	11-10	13.7
84037	11-7.5	—	83283	11-10	14.6
84044	11-7.5	—	83297	11-10	14.2
84052	11-7.5	—	83305	11-10	13.3
84058	11-7.5	—	83311	11-10	13.7
84065	11-7.5	—	83318	11-10	14.8
84072	11-7.5	—	83325	11-10	13.2
84079	11-7.5	—	83332	11-10	14.4
84086	11-7.5	—	83339	11-10	15.3
84093	11-7.5	—	83346	11-10	14.8
84100	11-7.5	15.5	83353	11-10	13.6
84107	11-7.5	14.8	83361	11-10	14.2
84114	11-7.5	14.6	84003	11-10	14.0
84121	11-7.5	15.3	84009	11-10	14.1
84128	11-7.5	15.7	84016	11-10	14.3
84135	11-7.5	14.4	84030	11-10	—
84142	11-7.5	15.3	84037	11-10	—
84151	11-7.5	—	84044	11-10	—
84156	11-7.5	—	84052	11-10	—
84163	11-7.5	—	84058	11-10	—
84170	11-7.5	—	84065	11-10	—
84177	11-7.5	—	84072	11-10	—
84184	11-7.5	—	84079	11-10	—
84191	11-7.5	—	84086	11-10	—
84198	11-7.5	15.1	84093	11-10	—
84205	11-7.5	14.7	84100	11-10	14.5

Table B1, Type: NP, continued.

Julian date	Sample# ^a	Reading (% VM)	Julian date	Sample# ^a	Reading (% VM)
84107	11-10	13.7	83361	11-12.5	14.6
84114	11-10	13.9	84003	11-12.5	15.8
84121	11-10	13.4	84009	11-12.5	14.5
84128	11-10	14.0	84016	11-12.5	15.0
84135	11-10	13.6	84030	11-12.5	—
84142	11-10	13.3	84037	11-12.5	—
84151	11-10	—	84044	11-12.5	—
84156	11-10	—	84052	11-12.5	—
84163	11-10	—	84058	11-12.5	—
84170	11-10	—	84065	11-12.5	—
84177	11-10	—	84072	11-12.5	—
84184	11-10	—	84079	11-12.5	—
84191	11-10	—	84086	11-12.5	—
84198	11-10	13.2	84093	11-12.5	—
84205	11-10	14.4	84100	11-12.5	14.9
84212	11-10	—	84107	11-12.5	15.1
84219	11-10	13.1	84114	11-12.5	13.9
84226	11-10	11.1	84121	11-12.5	14.8
83192	11-12.5	14.6	84128	11-12.5	15.1
83200	11-12.5	14.7	84135	11-12.5	15.1
83206	11-12.5	15.5	84142	11-12.5	13.8
83213	11-12.5	14.5	84151	11-12.5	—
83220	11-12.5	15.0	84156	11-12.5	—
83227	11-12.5	14.8	84163	11-12.5	—
83241	11-12.5	14.5	84170	11-12.5	—
83250	11-12.5	15.1	84177	11-12.5	—
83255	11-12.5	14.4	84184	11-12.5	—
83262	11-12.5	14.0	84191	11-12.5	—
83271	11-12.5	14.8	84198	11-12.5	14.7
83283	11-12.5	14.2	84205	11-12.5	14.6
83297	11-12.5	15.4	84212	11-12.5	—
83305	11-12.5	14.3	84219	11-12.5	14.1
83311	11-12.5	14.2	84226	11-12.5	12.5
83318	11-12.5	15.6	83192	11-15	14.0
83325	11-12.5	14.5	83200	11-15	14.2
83332	11-12.5	14.3	83206	11-15	13.5
83339	11-12.5	14.6	83213	11-15	13.0
83346	11-12.5	15.0	83220	11-15	13.8
83353	11-12.5	14.6	83227	11-15	13.3

Table B1, Type: NP, continued.

Julian date	Sample# ^a	Reading (%VM)	Julian date	Sample# ^a	Reading (%VM)
83241	11-15	14.4	84065	11-15	—
83250	11-15	14.7	84072	11-15	—
83255	11-15	14.1	84079	11-15	—
83262	11-15	13.9	84086	11-15	—
83271	11-15	13.9	84093	11-15	—
83283	11-15	14.5	84100	11-15	—
83297	11-15	14.4	84107	11-15	13.6
83305	11-15	14.5	84114	11-15	13.7
83311	11-15	14.4	84121	11-15	13.3
83318	11-15	13.9	84128	11-15	13.8
83325	11-15	13.8	84135	11-15	12.9
83332	11-15	13.7	84142	11-15	13.1
83339	11-15	13.3	84151	11-15	—
83346	11-15	12.9	84156	11-15	—
83353	11-15	13.7	84163	11-15	—
83361	11-15	14.1	84170	11-15	—
84003	11-15	13.0	84177	11-15	—
84009	11-15	13.9	84184	11-15	—
84016	11-15	13.7	84191	11-15	—
84030	11-15	—	84198	11-15	13.1
84037	11-15	—	84205	11-15	13.3
84044	11-15	—	84212	11-15	—
84052	11-15	—	84219	11-15	12.5
84058	11-15	—	84226	11-15	10.9

Table B1: continued.

(c) Type: T

Julian date	Sample# ^a	Reading (Ω)	Julian date	Sample# ^a	Reading (Ω)
83192	1,6-0-3	7	84086	1,6-0-3	0
83200	1,6-0-3	8	84093	1,6-0-3	0
83206	1,6-0-3	9	84100	1,6-0-3	0
83213	1,6-0-3	8	84107	1,6-0-3	0
83220	1,6-0-3	8	84114	1,6-0-3	0
83227	1,6-0-3	8	84121	1,6-0-3	0
83241	1,6-0-3	7	84128	1,6-0-3	0
83250	1,6-0-3	8	84135	1,6-0-3	0
83255	1,6-0-3	8	84142	1,6-0-3	0
83262	1,6-0-3	8	84151	1,6-0-3	0
83271	1,6-0-3	7	84156	1,6-0-3	0
83283	1,6-0-3	7	84163	1,6-0-3	0
83297	1,6-0-3	7	84170	1,6-0-3	4
83305	1,6-0-3	8	84177	1,6-0-3	3
83311	1,6-0-3	7	84184	1,6-0-3	4
83318	1,6-0-3	7	84191	1,6-0-3	4
83325	1,6-0-3	8	84198	1,6-0-3	3
83332	1,6-0-3	0	84205	1,6-0-3	4
83339	1,6-0-3	—	84212	1,6-0-3	—
83346	1,6-0-3	—	84219	1,6-0-3	4
83353	1,6-0-3	—	84226	1,6-0-3	3
83361	1,6-0-3	—	83192	2,7-1.5-5	5
84003	1,6-0-3	—	83200	2,7-1.5-5	7
84009	1,6-0-3	—	83206	2,7-1.5-5	8
84016	1,6-0-3	—	83213	2,7-1.5-5	9
84030	1,6-0-3	—	83220	2,7-1.5-5	8
84037	1,6-0-3	—	83227	2,7-1.5-5	9
84044	1,6-0-3	—	83241	2,7-1.5-5	6
84052	1,6-0-3	—	83250	2,7-1.5-5	6
84058	1,6-0-3	—	83255	2,7-1.5-5	7
84065	1,6-0-3	—	83262	2,7-1.5-5	8
84072	1,6-0-3	—	83271	2,7-1.5-5	8
84079	1,6-0-3	0	83283	2,7-1.5-5	8

^aFinal two numbers of the Sample # identify the location and depth of the instrument. See Sec. 3 and references cited therein for details of locations and descriptions.

Table B1, Type: T, continued.

Julian date	Sample# ^a	Reading (cbar)	Julian date	Sample# ^a	Reading (cbar)
83297	2,7-1.5-5	8	84212	2,7-1.5-5	—
83305	2,7-1.5-5	9	84219	2,7-1.5-5	3
83311	2,7-1.5-5	9	84226	2,7-1.5-5	4
83318	2,7-1.5-5	8	83200	3,8-5-3	22
83325	2,7-1.5-5	0	83206	3,8-5-3	24
83332	2,7-1.5-5	0	83213	3,8-5-3	29
83339	2,7-1.5-5	—	83220	3,8-5-3	28
83346	2,7-1.5-5	—	83227	3,8-5-3	27
83353	2,7-1.5-5	—	83241	3,8-5-3	13
83361	2,7-1.5-5	—	83250	3,8-5-3	13
84003	2,7-1.5-5	—	83255	3,8-5-3	15
84009	2,7-1.5-5	—	83262	3,8-5-3	16
84016	2,7-1.5-5	—	83271	3,8-5-3	16
84030	2,7-1.5-5	—	83283	3,8-5-3	14
84037	2,7-1.5-5	—	83297	3,8-5-3	16
84044	2,7-1.5-5	0	83305	3,8-5-3	15
84052	2,7-1.5-5	—	83311	3,8-5-3	15
84058	2,7-1.5-5	—	83318	3,8-5-3	15
84065	2,7-1.5-5	—	83325	3,8-5-3	16
84072	2,7-1.5-5	0	83332	3,8-5-3	0
84079	2,7-1.5-5	0	83339	3,8-5-3	—
84086	2,7-1.5-5	1	83346	3,8-5-3	—
84093	2,7-1.5-5	0	83353	3,8-5-3	—
84100	2,7-1.5-5	0	83361	3,8-5-3	—
84107	2,7-1.5-5	0	84003	3,8-5-3	—
84114	2,7-1.5-5	0	84009	3,8-5-3	—
84121	2,7-1.5-5	0	84016	3,8-5-3	—
84128	2,7-1.5-5	0	84030	3,8-5-3	—
84135	2,7-1.5-5	0	84037	3,8-5-3	—
84142	2,7-1.5-5	0	84044	3,8-5-3	0
84151	2,7-1.5-5	0	84052	3,8-5-3	—
84156	2,7-1.5-5	0	84058	3,8-5-3	0
84163	2,7-1.5-5	0	84065	3,8-5-3	0
84170	2,7-1.5-5	5	84072	3,8-5-3	0
84177	2,7-1.5-5	5	84079	3,8-5-3	0
84184	2,7-1.5-5	5	84086	3,8-5-3	0
84191	2,7-1.5-5	4	84093	3,8-5-3	0
84198	2,7-1.5-5	4	84100	3,8-5-3	0
84205	2,7-1.5-5	3	84107	3,8-5-3	1

Table B1, Type: T, continued.

Julian date	Sample# ^a	Reading (cbar)	Julian date	Sample# ^a	Reading (cbar)
84114	3,8-5-3	1	84003	4,9-6-5	—
84121	3,8-5-3	1	84009	4,9-6-5	—
84128	3,8-5-3	2	84016	4,9-6-5	—
84135	3,8-5-3	3	84030	4,9-6-5	—
84142	3,8-5-3	3	84037	4,9-6-5	—
84151	3,8-5-3	6	84044	4,9-6-5	4
84156	3,8-5-3	6	84052	4,9-6-5	4
84163	3,8-5-3	6	84058	4,9-6-5	5
84170	3,8-5-3	15	84065	4,9-6-5	6
84177	3,8-5-3	19	84072	4,9-6-5	7
84184	3,8-5-3	20	84079	4,9-6-5	8
84191	3,8-5-3	15	84086	4,9-6-5	11
84198	3,8-5-3	17	84093	4,9-6-5	12
84205	3,8-5-3	20	84100	4,9-6-5	14
84212	3,8-5-3	—	84107	4,9-6-5	14
84219	3,8-5-3	8	84114	4,9-6-5	9
84226	3,8-5-3	8	84121	4,9-6-5	16
83192	4,9-6-5	16	84128	4,9-6-5	15
83200	4,9-6-5	15	84135	4,9-6-5	16
83206	4,9-6-5	19	84142	4,9-6-5	14
83213	4,9-6-5	20	84151	4,9-6-5	16
83220	4,9-6-5	20	84156	4,9-6-5	16
83227	4,9-6-5	20	84163	4,9-6-5	16
83241	4,9-6-5	13	84170	4,9-6-5	15
83250	4,9-6-5	11	84177	4,9-6-5	15
83255	4,9-6-5	12	84184	4,9-6-5	13
83262	4,9-6-5	14	84191	4,9-6-5	14
83271	4,9-6-5	15	84198	4,9-6-5	16
83283	4,9-6-5	15	84205	4,9-6-5	19
83297	4,9-6-5	15	84212	4,9-6-5	—
83305	4,9-6-5	15	84219	4,9-6-5	15
83311	4,9-6-5	16	84226	4,9-6-5	12
83318	4,9-6-5	16	83192	5,10-11-10	6
83325	4,9-6-5	17	83200	5,10-11-10	6
83332	4,9-6-5	0	83206	5,10-11-10	5
83339	4,9-6-5	—	83213	5,10-11-10	11
83346	4,9-6-5	—	83220	5,10-11-10	8
83353	4,9-6-5	—	83227	5,10-11-10	11
83361	4,9-6-5	—	83241	5,10-11-10	6

Table B1, Type: T, continued.

Julian date	Sample# ^a	Reading (cbar)
83250	5,10-11-10	6
83255	5,10-11-10	9
83262	5,10-11-10	10
83271	5,10-11-10	12
83283	5,10-11-10	11
83297	5,10-11-10	10
83305	5,10-11-10	12
83311	5,10-11-10	13
83318	5,10-11-10	13
83325	5,10-11-10	14
83332	5,10-11-10	0
83339	5,10-11-10	—
83346	5,10-11-10	—
83353	5,10-11-10	—
83361	5,10-11-10	—
84003	5,10-11-10	—
84009	5,10-11-10	—
84016	5,10-11-10	—
84030	5,10-11-10	—
84037	5,10-11-10	—
84044	5,10-11-10	0
84052	5,10-11-10	0
84058	5,10-11-10	0
84065	5,10-11-10	0

Julian date	Sample# ^a	Reading (cbar)
84072	5,10-11-10	0
84079	5,10-11-10	1
84086	5,10-11-10	2
84093	5,10-11-10	2
84100	5,10-11-10	2
84107	5,10-11-10	2
84114	5,10-11-10	1
84121	5,10-11-10	3
84128	5,10-11-10	4
84135	5,10-11-10	4
84142	5,10-11-10	3
84151	5,10-11-10	5
84156	5,10-11-10	4
84163	5,10-11-10	4
84170	5,10-11-10	4
84177	5,10-11-10	4
84184	5,10-11-10	2
84191	5,10-11-10	4
84198	5,10-11-10	3
84205	5,10-11-10	1
84212	5,10-11-10	—
84219	5,10-11-10	2
84226	5,10-11-10	2

Table B2. Cambric tritium data.^a

(a) Type: L

Location	Date	dpm/mL	Error	Decay-corr	D(‰)
A6	83241	4206	ND	4227	—
	83250	4258	19	4279	—
	83255	4204	19	4225	—
	83272	4268	20	4307	—
	83272	4206	19	4244	—
	83292	4395	21	4435	—
	83297	4360	ND	4408	—
	83311	4090	41	4186	—
	83315	4366	ND	4415	—
	83318	3971	40	4064	—
	83325	4002	40	4096	—
	83332	3967	40	4060	—
	83339	3955	40	4048	—
	84003	3953	39	4046	-104
	84030	3771	38	3932	—
	84065	3694	37	3852	—
	84114	3881	39	4047	—
	84121	3832	38	3996	—
	84156	3898	20	4125	—
	84212	4032	20	4267	—
	84248	3437	17	3637	—
	84275	3443	17	3659	—
	84310	3120	31	3410	—
	84337	3095	31	3383	—
	85002	3068	31	3353	—
	85062	3121	31	3411	—
	85091	2802	28	3093	—
	85182	2870	29	3214	—
	85217	2697	27	—	—
	85248	2723	27	—	—
	85280	2753	27	—	—
	85308	2635	26	—	—
	85336	2464	24	—	—
	86006	2509	24	—	—

^a The results of LLNL tritium determinations are expressed in disintegrations per minute per milliliter of water (dpm/mL). Errors reported are in dpm/mL, and are two standard deviations based on counting statistics alone. DRI deuterium determinations are expressed in per mille relative to the Standard Mean Ocean Water (SMOW) standard.

Table B2, (a) Type L, continued.

Location	Date	dpm/mL	Error	Decay-corr	D(‰)
A6	86034	2531	24	—	—
	86062	2531	24	—	—
	86097	2420	24	—	—
	86125	1770	18	Note decrease	—
	86153	2260	9	—	—
	86203	2150	22	—	—
	86224	2120	21	—	—
	86253	2110	21	—	—
	86282	2090	20	—	—
C12	83241	5029	ND	5054	—
	83250	5010	21	5035	—
	83255	5048	21	5073	—
	83262	5151	22	5198	—
	83272	5029	20	5054	—
	83272	5047	21	5072	—
	83283	5181	23	5228	—
	83297	5228	ND	5286	—
	83311	4842	48	4956	—
	83325	4739	48	4851	—
	83332	4858	49	4972	—
	83339	4849	49	4963	—
	84030	4669	47	4869	—
	84065	4613	46	4810	—
	84121	4596	46	4792	—
	84156	3667	20	3880	—
	84248	4282	22	4531	—
	84275	4231	21	4496	—
	84310	4080	41	4459	—
	84337	3988	40	4358	—
	85002	3947	39	4314	—
	85035	3878	39	4238	—
	85062	3886	38	4247	—
	85091	3670	37	4051	—
	85154	3598	36	3982	—
	85217	3536	35	—	—
	85280	3419	33	—	—
	85308	3368	33	—	—
	86006	3108	31	—	—
	86034	3108	31	—	—

Table B2, (a) Type L, continued.

Location	Date	dpm/mL	Error	Decay-corr	D(%)
C12	86062	3041	30	—	—
	86097	2910	29	—	—
	86125	2840	28	—	—
	86203	2690	27	—	—
	86224	2600	26	—	—
	86253	2640	26	—	—
	86282	2550	26	—	—
D6	83255	353.8	3.3	356	—
	83262	494.9	4.1	504	—
	83271	374.4	3.4	378	—
	83297	430	ND	435	—
	83325	443	11	453	—
	83332	338	9	346	—
	84065	568	6	592	—
	84121	672	7	701	—
	84212	871.1	9.8	922	—
	84275	1150	10	1222	—
	84310	1100	11	1202	—
	84337	570.3	5.2	623	—
	85035	891.9	9.0	975	—
	85062	695.6	7.0	760	—
	85091	550.6	5.6	608	—
	85126	524	5	575	—
	85154	520	5	574	—
	86125	1700	—	15	—
E12	84212	852.7	9.8	903	—
	84310	787.9	7.9	861	—
	84337	801.7	8.1	876	—
	85002	864.6	8.7	945	—
	85035	893.8	9.0	977	—
	85062	1005	10	1098	—
	85091	921.5	9.3	1017	—
	85126	1178	12	1293	—
	85154	921	9	1017	—
	85182	955	10	1069	—
	85248	935	9	—	—
	85280	1137	—	11	—
	86006	1190	—	12	—
	86308	1061	—	11	—

Table B2, (a) Type L, continued.

Location	Date	dpm/mL	Error	Decay-corr	D(‰)
E12	86062	1361	—	14	—
	86125	1620	—	16	—
	86153	1600	—	6.4	—
	86224	1760	—	18	—
F5	85091	10.91	0.61	12.04	—
	85062	34.08	0.91	37.23	—
G11	84310	10.70	0.61	11.69	—
	85002	8.35	0.57	9.13	—
	85091	1.02	0.42	1.13	—
	85182	1.42	0.46	1.59	—
	85280	9.90	0.60	—	—
	85308	5.39	0.53	—	—
	86006	2.29	0.44	—	—
	86034	6.95	0.58	—	—
	86062	0.71	0.44	—	—
	86097	0.73	0.40	—	—
	86125	1.01	0.42	—	—
	86153	1.41	0.44	—	—
L-1	83332	2270	25	2323	—
	83339	2267	25	2320	—
	84003	3000	30	3071	—
	84065	2929	29	3054	—
	84307	2392	24	2614	—
	85035	2352	24	2570	—
	85091	2194	22	2422	—
	85126	2219	22	2436	—
	85154	2200	22	2429	—
	85182	2650	27	2968	—
	85280	2575	27	—	—
	85308	2453	24	—	—
	86062	2353	24	—	—
	86097	2400	24	—	—
	86125	2290	23	—	—
L-2	86153	2330	40	—	—
	83332	1294	18	1324	—
	83339	1391	19	1424	—
	84065	1993	20	2078	—

Table B2, (a) Type L, continued.

Location	Date	dpm/mL	Error	Decay-corr	D(‰)
L-2	84337	1841	18	2012	—
	85002	1640	16	1792	—
	85091	708.6	7.2	782	—
L-100	85203	2191	22	—	—
	85203	2674	27	—	—
	85217	2695	27	—	—
	85231	2704	27	—	—
	85238	2697	27	—	—
	85246	2711	27	—	—
	85252	2731	27	—	—
	85259	2686	27	—	—
	85273	2708	27	—	—
	85280	2731	27	—	—
	85287	2686	27	—	—
	85294	2691	27	—	—
	85301	2717	27	—	—
	85324	2597	27	—	—
	85329	2575	27	—	—
	85343	2553	27	—	—
	85350	2553	27	—	—
	85357	2531	24	—	—
	85364	2553	27	—	—
	86013	2575	27	—	—
	86021	2553	27	—	—
	86027	2575	27	—	—
	86041	2553	27	—	—
	86049	2575	27	—	—
	86055	2553	27	—	—
	86069	2530	24	—	—
	86076	2571	11	—	—
	86081	2533	11	—	—
	86083	2522	11	—	—
	86084-1	2513	11	—	—
	86084-2	2544	11	—	—
	86085-1	2533	11	—	—
	86085-2	2546	11	—	—
	86086-1	2529	11	—	—
	86086-2	2524	11	—	—
	86087-1	2513	11	—	—

Table B2, (a) Type L, continued.

Location	Date	dpm/mL	Error	Decay-corr	D(%)
L-100	86087-2	2502	11	—	—
	86088	2529	11	—	—
	86089	2526	11	—	—
	86090-1	2522	11	—	—
	86090-2	2509	11	—	—
	86091	2540	11	—	—
	86092	2511	11	—	—
	86093-1	2546	11	—	—
	86093-2	2517	11	—	—
	86094	2533	11	—	—
	86097-1	2513	11	—	—
	86097-2	2515	11	—	—
	86098-1	2509	11	—	—
	86098-2	2517	11	—	—
	86100	2498	11	—	—
	86101-1	2480	11	—	—
	86101-2	2493	11	—	—
	86104-1	2493	11	—	—
	86104-2	2462	11	—	—
	86105	2511	11	—	—
	86107-1	2506	11	—	—
	86107-2	2460	11	—	—
	86111-1	2473	11	—	—
	86111-2	2504	11	—	—
	86113	2364	11	—	—
	86114	2389	11	—	—
	86118-1	2391	11	—	—
	86118-2	2364	11	—	—
	86121-1	2367	11	—	—
	86121-2	2378	11	—	—
	86125-1	2373	11	—	—
	86125-2	2362	11	—	—
	86147-1	2350	9.1	—	—
	86147-2	2350	9.1	—	—
	86149	2310	9.3	—	—
	86153-1	2330	9.3	—	—
	86153-2	2330	9.3	—	—
	86160-1	2330	9.3	—	—
	86160-2	2330	9.3	—	—

Table B2, (a) Type L, continued.

Location	Date	dpm/mL	Error	Decay-corr	D(‰)
L-100	86174	2310	9.1	—	—
	86177	2310	9.3	—	—
	86188	2290	9.1	—	—
	86203	2260	9.1	—	—
	86216	2210	22	—	—
	86224	2190	22	—	—
	86246	2200	22	—	—
	86253	2200	22	—	—
	86275	2160	22	—	—
	86282	2140	22	—	—

Table B2, (b) Type: CCW

Location	Date	dpm/mL	Error	Decay-corr	D(‰)
OUT	83154	4427	20	—	—
F-1	83154	4455	20	4455	—
F-2	83154	4445	20	4445	—
F-1	83271	4186	20	4207	-101
F-2	83271	4170	19	4191	-103
	84030	3754	38	3914	—
	84065	3680	37	3837	—
	84114	3594	36	3748	—
	84121	3557	36	3709	—
	84156	3863	20	4088	—
	84212	3759	20	3978	—
	84220	3756	20	3975	—
	84220	3770	20	3989	—
	84221	3754	20	3972	—
F-3	84221	3814	20	4036	—
	84248	3279	16	3499	—
	84282	3073	31	3297	—
	84310	3048	30	3270	—
	84337	3021	30	3287	—
	85002	2957	30	3218	—
	85035	2957	29	3232	—
	85062	2916	29	3187	—
	85091	2877	29	3175	—

Table B2, (b) Type: CCW, continued.

Location	Date	dpm/mL	Error	Decay-corr	D(%)
	85126	2730	27	3040	—
	85154	2693	27	2999	—
	85182	2684	27	3017	—
	85217	2624	26	—	—
	85248	2561	26	—	—
	85280	2509	24	—	—
	85308	2494	25	—	—
	86034	2464	24	—	—
	86062	2398	24	—	—
	86084	2313	11	—	—
	86085-1	2311	11	—	—
	86085-2	2307	11	—	—
	86085-3	2291	11	—	—
	86085-4	2249	11	—	—
	86087	2293	11	—	—
	86097	2295	11	—	—
	86105	2253	11	—	—
	86107	2269	11	—	—
	86111	2278	11	—	—
	86113	2207	11	—	—
	86114	2202	11	—	—
	86116	2267	11	—	—
	86118	2204	11	—	—
	86121	2213	11	—	—
	86125	2204	11	—	—
	86147	2200	8.9	—	—
	86149	2200	8.9	—	—
	86153	2200	8.9	—	—
	86160	2170	8.7	—	—
	86168	2160	8.7	—	—
	86174	2160	8.7	—	—
	86177	2170	8.7	—	—
	86188	2160	8.7	—	—
	86203	2150	8.7	—	—
	86216	2100	9.4	—	—
	86224	2080	21	—	—
	86246	2020	20	—	—
	86253	2050	20	—	—
	86275	2030	20	—	—
	86282	2020	20	—	—

Table B2, (c) Type: LANL CCW

Date	dpm/mL	Decay-corr
83131	4375	4292
83145	4356	4273
83154	4294	4234
83154	4455	4393
83160	4296	4236
83168	4264	4204
83173	4268	4208
83181	4238	4179
83188	4215	4177
83195	4161	4124
83202	4183	4145
83215	4113	4092
83223	4093	4073
83229	4196	4175
83237	4249	4228
83245	4210	4210
83250	4232	4232
83258	4064	4064
83265	4051	4051
83271	4020	4020
83271	4186	4186
83287	4026	4046
83300	3965	3985
83306	4028	4065

Date	dpm/mL	Decay-corr
83314	4021	4058
83320	4019	4055
83326	3895	3930
83335	3857	3912
83343	3854	3909
83348	3857	3912
83356	3812	3866
83363	3784	3838
84012	3760	3833
84018	3742	3814
84026	3764	3837
84033	3768	3857
84040	3769	3858
84047	3732	3820
84054	3712	3799
84061	3710	3817
84069	3691	3797
84156	3863	4011
84212	3759	3920
84220	3756	3937
84221	3754	3935
84230	3241	3430
84263	3197	3397

Table B2, (d) Type: FDSW

Location	Date	dpm/mL	Error
4-5	83153	5056	23
10-5	83153	189	2
20-5	83153	1.03	0.04
0-7	83153	4141	21
0-3	83153	4021	20
0-5	83153	4074	20
4-10	83153	4922	22
10-10	83153	145.6	2.0
20-10	83153	4.7	0.13
11-15	83153	1984	12
FF-1,1	83145	24.0	1.1
FF-1,2	83145	22.3	1.0

Location	Date	dpm/mL	Error
FF-1,3	83145	14.2	0.7
FF-1,4	83145	9.4	0.5
FF-1,5	83145	6.1	0.4
FF-1,7	83145	4.1	0.2
L-100-6.5	85123	2758	28
L-100-11	85123	2826	28
L-100-21.5	85123	2787	28
L-100-31.5	85123	2745	27
L-100-42	85123	2768	28
L-100-57	85123	2789	28
L-100-71.5	85123	2780	28
L-100-100	85123	2764	28

Table B2, (e) Type: RW

Location	Date	dpm/mL	Error
plot	84267	1.40	0.45
plot	84282	10.0	0.7
plot	85254	5.65	0.56

Table B2, (f) Type: SAW

Location	Date	dpm/mL	Error
A	—	4.03	0.48
B	—	10.2	0.6
A	85093	12.7	0.62
B	85093	4.60	0.49
B	85224	4.44	0.48
A	86013	22.9	0.76
B	86013	30.4	0.84

Table B2, (g) Type: CPW

Location	Date	dpm/mL	Error	D(‰)
EEOL	83154	2850	16	—
	83271	2753	15	-88
SEOL	83154	3749	18	—
	83271	3179	17	-93
SOL	83154	4177	19	—
	83271	2561	15	-82
W-1	84039	3786	38	—
W-2	84039	3789	38	—
	84220	3530	20	—
W-3	84039	3807	38	—
	84220	3732	20	—
W-4	84039	3787	38	—
	84220	3743	20	—
W-5	84039	3716	37	—
W-6	84039	3537	36	—

Location	Date	dpm/mL	Error	D(‰)
W-7	84039	3541	35	—
W-8	84039	3477	35	—
W-9	84039	3780	38	—
W-10	84039	3787	38	—
	84220	3623	20	—
W-11	84039	3755	38	—
	84220	3561	20	—
W-12	84039	3776	38	—
	84220	3712	20	—
W-13	84039	3800	38	—
	84220	3403	20	—
W-14	84039	3824	38	—
	84220	3712	20	—
W-15	84039	3802	38	—

Table B2, (h) Type: AMS

Location	Date	dpm/mL	Error	D(‰)
Ditch	84220	748	9	-108
Ditch	84220	333	6	-112
Remote	84220	30.9	2.9	-108
Remote	84220	62.8	3.3	-82

Table B2, (i) Type: TPS^b

Location	dpm/mL	Error
DRI01	1065	7
DRI02	1543	10
DRI03	1703	11
DRI04	838	8
DRI05	1363	10
DRI06	1140	7
DRI07	614	6
DRI08	713	8
DRI09	1337	11
DRI10	1395	10
DRI11	972	8
DRI12	935	8
DRI13	1318	10
DRI14	1138	7
DRI15	700	7
DRI16	602	10
DRI17	1447	10
DRI18	1853	12
DRI19	855	8
DRI20	828	8
DRI21	258	4
DRI22	314	5
DRI23	748	8
DRI24	640	6
DRI25	853	8
DRI26	770	13
DRI27	1045	20
DRI28	1125	9
DRI29	2697	55

Location	dpm/mL	Error
DRI31	904	8
DRI32	865	9
DRI33	1038	17
DRI34	208014	
DRI35	919	8
DRI36	936	8
DRI37	1226	20
DRI38	1608	11
DRI39	42	1
DRI40	52	2
DRI41	27	1
DRI42	25	1
DRI43	1176	9
DRI44	732	7
DRI45	62	2
DRI46	58	2
DRI47	34	1
DRI48	34	1
DRI49	22	1
DRI50	26	1
DRI51	2435	14
DRI52	2272	13
DRI53	950	8
DRI54	974	8
DRI55	2383	14
DRI56	2183	13
DRI57	1368	10
DRI58	1769	11

^b See Table B3 for descriptions of these samples.

Table B2, (j) Type: FDPW

Code/Location	Date	Time	dpm/mL	Error	D(%)
F1-1	83154	—	2531	15	—
F1-2	83154	—	2898	16	—
EOL-1	83154	—	2136	12	—
MC01/1-4	83271	0730	1336	8	-85
MC02/5-6	83271	0730	1776	10	-90
MC03/7-8	83271	0730	911	6	-78
MC04/9-10	83271	0730	2030	12	-75
MC05/11-12	83271	0730	829	5	-92
MC06/21-22	83271	—	331	3	-61
MC07/23-24	83271	—	669	5	-80
MC08/25-26	83271	—	1217	7	-62
MC09/27-28	83271	—	1422	8	-69
MC10/37-38	83271	—	1969	11	-78
MC11/39-40	83271	—	41.2	1.1	-58
MC12/41-42	83271	—	17.4	0.4	-63
MC13/51-52	83271	1200	2597	15	-77
MC14/53-54	83271	1200	1306	8	-63
MC15/55-56	83271	1200	2777	16	-73
MC16/57-58	83271	1200	1169	7	-78
1/CT	84220	0800	1852	13	-2
2/SC	84220	0800	2646	18	-67
3/CT6-7	84220	0830	1645	13	0
4/CT3-4	84220	0830	2635	18	-49
5/CT1-2	84220	0830	3337	20	-119
6/CT	84220	1200	2804	18	+35
7/SC	84220	1200	3481	20	-67
8/CT	84220	1600	3374	20	+21
9/SC	84220	1600	3576	20	-85
10/CT6-7	84220	1630	3514	20	+19
11/CT3-4	84220	1630	3787	20	-80
12/CT1-2	84220	1630	3858	20	-97

Table B3. Toluene Plant Samples (TPS).

DRI Sample #	Sample (28 Sept 84)
1-4*	Methods-comparisons sample: taken near dawn using <i>Conyza coulteri</i>
5-12*	0700 h diurnal C ₃ - C ₄ comparisons
(5-6)	<i>Conyza coulteri</i> (leafy C ₃ dicot weed; common in moist places)
(7-8)	<i>Salsola iberica</i> (C ₄ dicot weed; most common in open waste places)
(9-10)	<i>Typha domingensis</i> (C ₃ monocot wetland plant; cattail)
(11-12)	<i>Euphorbia albomarginata</i> (mat-like C ₄ perennial herb)
13-20	0830 h diurnal
(13-14)	<i>Conyza coulteri</i>
(15-16)	<i>Salsola iberica</i>
(17-18)	<i>Typha domingensis</i>
(19-20)	<i>Euphorbia albomarginata</i>
21-28*	Near Frenchman Flat berm, from marsh ca. 200 m from end of canal
(21-22)	<i>Typha domingensis</i> , 200 m N of canal line, 0930 h
(23-24)	<i>Tamarix pentandra</i> (C ₃ tree; salt cedar), same as above
(25-26)	<i>Typha domingensis</i> , in line with canal, 1000 h
(27-28)	<i>Tamarix pentandra</i> , same as above
29-36	1030 h diurnal
(29-30)	<i>Conyza coulteri</i>
(31-32)	<i>Salsola iberica</i>
(33-34)	<i>Typha domingensis</i>
(35-36)	<i>Euphorbia albomarginata</i>

* Duplicate samples taken by LLNL.

Table B3. Continued.

DRI	
Sample #	Sample (28 Sept 84)
37-42 ^a	C ₃ species transect from berm; <i>Sphaeralcea coulteri</i> (globe mallow), a leafy herbaceous "roadside" weed used in transect
(37-38)	Next to berm (but outside it), a 50 x 50 cm plant
(39-40)	6-7 m from berm (berm side of road), population of small plants
(41-42)	12 m from berm (across road), population of small plants
43-50	C ₄ species transect from berm; <i>Atriplex canescens</i> (four-wing saltbush) used in transect
(43-44)	Inside berm, 2 shrubs each 60 x 40 cm
(45-46)	6-7 m from berm (berm side of road), 40 x 60 cm bush
(47-48)	12 m from berm (across road), 2 bushes each 50 x 50 cm
(49-50)	30 m from berm, two large bushes each 100 x 100 cm
51-58 ^a	1200 h diurnal
(51-52)	<i>Conyza coulteri</i>
(53-54)	<i>Salsola iberica</i>
(55-56)	<i>Typha domingensis</i>
(57-58)	<i>Euphorbia albomarginata</i>

^a Duplicate samples taken by LLNL.

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LABORATORY AND FIELD STUDIES
RELATED TO THE RADIONUCLIDE MIGRATION PROJECT

October 1, 1985—September 30, 1986

Compiled and Edited by Joseph L. Thompson

ABSTRACT

In this report we describe the work done at Los Alamos in support of the Radionuclide Migration project during fiscal year 1986. We have continued to monitor the transport of tritium and ^{85}Kr from the Cambria explosion zone to the satellite well, which is pumped at 600 gal./min. Corresponding movement of cationic radionuclides such as ^{137}Cs and ^{90}Sr has not yet been observed after 12 yr of pumping, nor have we seen evidence that these strongly sorbing ions move in conjunction with colloids. We have analyzed more data from the Cheshire study site but have not resolved the uncertainties regarding the distribution and movement of radioactive materials at this location. Our attempts to improve our analytical capability for ^{36}Cl and ^{99}Tc have resulted in some progress. Similarly, we have increased our understanding of radionuclide transport phenomena such as channeling in fracture flow and anion exclusion in zeolites and clays. A sample exchange with Lawrence Livermore National Laboratory has helped us identify critical steps in our procedures for collecting and analyzing large-volume water samples. We have surveyed potential sites on Pahute Mesa at the Nevada Test Site for future radionuclide migration studies and conclude that there are none other than Cheshire presently available, and none are likely to be created in the near future. The Laboratory has engaged recently in radionuclide migration studies sponsored by our weapons program; we review this work in an appendix to the annual report.

I. INTRODUCTION

The Radionuclide Migration (RNM) project was initiated in 1973 to study the movement of radionuclides at the Nevada Test Site (NTS). The agencies involved in this project seek to understand how radioactive materials associated with underground nuclear tests may migrate through the groundwater system. The principal organizations involved in the RNM work are Los Alamos National Laboratory (LANL), Lawrence Livermore National Laboratory (LLNL), the US Geological Survey, the Desert Research Institute, and the Nevada Operations Office of the US Department of Energy; they are assisted by various support organizations at the NTS. Contributions to the program by LANL are generally in the areas of radionuclide detection, geochemistry, and radionuclide interactions with geologic media. Our principal study sites at the NTS are in Area 5 (the Cambric site) and in Area 20 (the Cheshire site). We have worked at the Cambric site since 1974 and have learned a good deal about the behavior of radionuclides in tuffaceous alluvium. The Cheshire site has been studied for a shorter time. It is located in brecciated rhyolite and is more complex geologically and hydrologically than the Cambric site is. We also examine other nuclear test sites as the opportunity arises; one such study is described in the Appendix to this report. Our goal is to develop an understanding of the basic phenomenology of radionuclide transport in the underground environment at the NTS. The opportunity to make field observations is an invaluable adjunct to our laboratory experimental programs, allowing us to verify predictions and validate mathematical models. Much of the RNM work is also of interest to the Nevada Nuclear Waste Storage Investigation project, and many Los Alamos personnel contribute to both programs.

In this report we review our accomplishments in the RNM project during the past year. New data are presented and often the entire data base is reproduced to show trends. Some subjects have been treated in detail in previous reports and in such cases references are given to the earlier documents. This year we have adopted $\mu\text{Ci}/\text{ml}$ as our standard unit of concentration, whereas atoms/ml was often used in the past, so some tables and figures will appear altered from previous reports.

II. CAMBRIC STUDIES

The Cambric site is the location of a 0.75-kt nuclear test conducted in 1965 at a depth of 294 m in tuffaceous alluvium in Frenchman Flat at the NTS. The detonation point was 73 m below the water table, but there is little water movement at this depth, and when we re-entered the cavity and chimney regions in 1974 we found that the tritium and fission products associated with the test shot were still there. After we measured the distribution of the radionuclides between the soil and water, we began in 1974 testing to see what radioactive materials would move as the groundwater moved. We pumped water from a satellite well (RNM-2S) 91 m from the cavity, first at 300 gal./min and after 2 yr at 600 gal./min. We have continued to monitor the concentrations of radionuclides in the effluent water from RNM-2S and in the RNM-1 water. The work at the Cambric site is described in several articles¹⁻³ and annual reports.⁴⁻⁹ We present here new data acquired during the past year and discuss its relation to the existing data set.

A. Sampling and Counting Methodologies

Because our water samples from Cambric and other sites contain very small concentrations of radionuclides, we have developed sampling and counting protocols that enable us to measure the species of interest at or below the pCi/ml level. We described our procedures in some detail in our annual report for last year,⁹ so here we will only indicate the general method used and the level of detection routinely achieved.

Tritium analysis of pumped or bailed water samples is done by liquid scintillation counting. Our normal measurement limits are $\sim 2 \times 10^{-6}$ $\mu\text{Ci/ml}$ of water. Water samples containing ^{85}Kr are collected in gas-tight "pressurized tubes" and the ^{85}Kr is separated using a vacuum line. The ^{85}Kr is beta counted with a proportional counter over a period of several weeks. The detection limit for ^{85}Kr is $\sim 1 \times 10^{-8}$ $\mu\text{Ci/ml}$ of the water sample. We frequently measure 208- ℓ (55-gal.) water samples for the presence of gamma-emitting radionuclides. We concentrate these samples using a glass still, then evaporate them to dryness, and count them with a germanium detector. With this procedure we can detect ^{137}Cs at concentrations down to 4×10^{-11} $\mu\text{Ci/ml}$. We may separate ^{90}Sr or ^{137}Cs from water samples by using radiochemical procedures. If we start with 2- ℓ samples, our

measurement levels are ~ 1 to 2×10^{-8} $\mu\text{Ci}/\text{mL}$. We can chemically separate plutonium and alpha count the purified product. The counting limit for ^{239}Pu is $\sim 2 \times 10^{-11}$ $\mu\text{Ci}/\text{mL}$.

Nonroutine sample analyses for radionuclides such as ^{36}Cl , ^{99}Tc , and ^{129}I require more exotic procedures such as tandem-accelerator mass spectrometry, neutron activation analysis, and isotope dilution mass spectrometry. The use of such techniques is described for particular radionuclides in this and previous annual reports.

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

We have continued to monitor the decreasing levels of tritium in the effluent from the RNM-2S well. The latest data are given in Table I and the complete data set is displayed in Figs. 1 and 2. The shape of the tail of the elution curve will enable us to validate various models proposed to explain the movement of tritium between the RNM-1 and RNM-2S wells.^{5,8} The pump at RNM-2S was off from 12/05/85 to 01/21/86; the unusually large scatter in the data at about this time may be related to that fact. Through the end of September 1986, $\sim 75\%$ of the initial tritium inventory has been pumped out of RNM-2S.

C. Fission Product Migration (S. Maestas, A. E. Ogard, A. J. Mitchell, F. O. Lawrence, J. Drake, D. W. Efurd, J. D. Gallagher, and J. L. Thompson)

We have listed in Table II the ^{85}Kr concentrations measured in RNM-2S water samples during the past year and in Fig. 3 have displayed the entire elution curve for this radionuclide. Although the elution pattern is generally similar, it is apparent that krypton is eluted somewhat after tritium. The ratio of ^{85}Kr /tritium as given in Table II and displayed in Fig. 4 shows a slowly rising trend but is significantly less than the source term ratio of 1.22×10^{-4} .

At RNM-1 the tritium and ^{85}Kr concentrations continue to decrease, as shown in Tables III and IV. However, the ^{85}Kr /tritium ratio remains higher than the source term ratio, again reflecting the fact that the krypton has not moved out of the cavity region as readily as has the tritium.

New data for ^{90}Sr and ^{137}Cs in RNM-1 water are given in Table V. We have included the complete data set for these radionuclides because we are using different units than in

TABLE I
TRITIUM ANALYSES OF RNM-2S WATER SAMPLES

Identification Number	Date Collected	Volume Pumped (10 ⁶ gals.)	Tritium ^a (10 ⁻³ μ Ci/ml)
G 1449	11/20/85	2837.3	1.06
G 1450	12/04/85	2847.0	0.94
b			
G 1451	01/22/86	2847.7	1.05
G 1452	01/28/86	2852.9	1.09
G 1453	02/04/86	2859.1	1.02
G 1454	02/10/86	2864.4	1.01
G 1455	02/19/86	2872.3	1.01
G 1456	02/27/86	2879.5	1.00
G 1457	04/03/86	2910.1	0.96
G 1458	05/22/86	2952.8	0.95
G 1459	06/20/86	2978.1	0.94
G 1460	07/23/86	3007.1	0.91
G 1461	08/19/86	3030.1	0.89
G 1462	09/27/86	3063.6	0.87

^aStandard deviation of counting data <1%.

^bNo pumping from 12/05/85 to 01/21/86; new pump installed 01/21/86.

previous years. The trend seen in recent years continues; namely the ⁹⁰Sr and ¹³⁷Cs concentrations are decreasing at a much slower rate than the concentration of the tritium. To date we have no evidence of either ⁹⁰Sr or ¹³⁷Cs in RNM-2S water. The large sorption ratios of these anions on tuffs, as measured in our laboratory,¹⁰ preclude their transport as dissolved species. We might anticipate that colloid-sized particles could carry either of these radionuclides over the 91-m distance between the two wells. However, our filtration studies of the RNM-2S water indicate that this water is very low in particulate content relative to other pumped water on the NTS. Apparently, the tuffaceous alluvium is a very effective filter, so colloid transport is not occurring to a measurable extent.

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

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

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

Identification Number	Date Collected	Volume Pumped (10^6 m^3)	^{85}Kr Concentration		$^{85}\text{Kr}/\text{Tritium}$	
			$10^{-3} \mu\text{Ci}/\text{m}\ell$	$10^{-7} \mu\text{Ci}/\text{m}\ell$	Atom Ratio ^b $\times 10^5$	Ratio ^c
434-1-85-019	11/20/85	10.740	3.32	2.67	7.00	0.57
434-1-85-021	12/04/85	10.777	3.01	2.42	7.00	0.57
434-1-86-001	01/24/86	10.787	3.38	1.60	4.11	0.34
434-1-86-003	02/19/86	10.873	3.24	2.54	6.82	0.56
434-1-86-005	03/19/86	10.966	3.17	2.32	6.36	0.52
434-1-86-007	04/03/86	11.014	3.13	2.46	6.84	0.56
434-1-86-009	05/22/86	11.176	3.10	2.46	6.91	0.57
434-1-86-013	07/08/86	11.333	3.01	2.30	6.65	0.54
434-1-86-015	08/05/86	11.424	2.95	2.42	7.14	0.58
434-1-86-017	09/03/86	11.521	2.94	2.40	7.10	0.58

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

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

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

Fig. 3. The ^{85}Kr concentration in water pumped from RNM-2S.

Fig. 4. Atom ratio of ^{85}Kr to tritium in water pumped from RNM-2S.

TABLE III
TRITIUM CONCENTRATION IN RNM-1 WATER (1986)

Identification Number	Volume Pumped (m ³)	Tritium Concentration (10 ⁻⁶ μCi/ml)	
		On Collection Date 5/7/86	Corrected to Cambric t ₀ 5/14/65
S-1	1.5	1.55	5.05
S-2	11.0	1.80	5.86
S-3	24.2	1.80	5.86
S-4	31.8	1.72	5.60
S-5	44.3	1.68	5.47
S-6	51.5	1.72	5.60
S-7	61.3	1.80	5.86
434-3-86-001 ^a	60.6	2.16	7.10
			Average 5.80

^aCollected in a gas-tight pressurized tube.

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

Zone	Date	Volume Pumped ^a (10 ⁶ m ³)	Concentration ^b		⁸⁵ Kr/Tritium	
			Tritium (μCi/ml)	⁸⁵ Kr (μCi/ml)	Atom Ratio ^c (× 10 ⁴)	Ratio ^d
Original-Zone IV	08/08/75	0.	1.5 × 10 ⁻¹	3.1 × 10 ⁻⁵	1.8	1.5
Original-Zone V	08/14/75	0.	3.8 × 10 ⁻²	6.1 × 10 ⁻⁶	1.4	1.2
Zone IV + V	10/04/77	1.17	3.2 × 10 ⁻³	3.4 × 10 ⁻⁵	92	75
Zone IV + V	11/30/77	1.34	2.0 × 10 ⁻³	2.7 × 10 ⁻⁶	12	10
Zone IV + V	09/04/79	3.50	2.6 × 10 ⁻⁴	2.5 × 10 ⁻⁷	8.5	7
Zone IV + V	10/05/81	5.89	1.4 × 10 ⁻⁴	8.3 × 10 ⁻⁸	5.0	4
Zone IV + V	04/01/82	6.48	7.7 × 10 ⁻⁵	3.0 × 10 ⁻⁸	3.3	3
Zone IV + V	04/06/83	7.67	3.3 × 10 ⁻⁵	1.3 × 10 ⁻⁸	3.4	3
Zone IV + V	04/24/84	8.91	2.2 × 10 ⁻⁵	1.0 × 10 ⁻⁸	4.1	3
Zone IV + V	04/10/85	10.0	1.3 × 10 ⁻⁵	1.2 × 10 ⁻⁸	7.8	6
Zone IV + V	05/07/86	11.1	5.8 × 10 ⁻⁶	2.1 × 10 ⁻⁹	3.2	3

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

^b Values corrected to Cambric zero time.

^c 1.81 × 10¹³ atoms ⁸⁵Kr = 1 μCi; 2.08 × 10¹³ atoms tritium = 1 μCi.

^d Measured ⁸⁵Kr/tritium divided by ⁸⁵Kr/tritium calculated for Cambric (1.22 × 10⁻⁴).

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

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

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

^b Values corrected to Cambrian zero time.

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

III. CHESHIRE STUDIES (J. L. Thompson, S. Maestas, F. O. Lawrence, G. W. Knobeloch, D. W. Efurud, J. Drake, and J. D. Gallagher)

As noted in the Introduction to this document, the Cheshire site is quite different from the Cambric site geologically, and it has hydraulic gradients that may cause movement of some of the radionuclides associated with the test. The test device was fired in the brecciated rhyolite of Pahute Mesa in February 1976 at a depth of 1174 m, ~544 m below the water table. We did limited sampling near the cavity region in late 1976; this effort was terminated when a pump became stuck in the re-entry hole. In 1983 and 1984 we collected water samples through perforations in the casing at a depth of about 1200 m. In 1985 we set a packer above 1200 m, made new perforations at 820 m, and collected more water samples. During the past year, we have analyzed a few water samples for tritium and ^{85}Kr and have boiled down and counted two 208- ℓ samples, one taken just before the 820-m perforations were made and one taken several months later. These analyses add to our data base but do not answer the questions concerning data interpretation that were discussed in our report from last year.⁹

Table VI contains tritium analyses results obtained since September 1985. These values, added to those in Table VI of Ref. 9 and Table IX of Ref. 8, complete our data set for tritium measurements for Cheshire samples. In Table VII we presented our complete data set for tritium and ^{85}Kr measured in pressurized samples from Cheshire. Finally, in Table VIII we list our most recent analyses of gamma-emitting radionuclides in evaporated 208- ℓ water samples. This latter group may be compared with the data in Table VIII of Ref. 9.

In general, the concentrations of all of the radionuclides sampled have decreased as water was pumped from the hole at Cheshire. This trend can be seen in Figs. 5 and 6. The surprising observation is that most species are present in about the same concentrations in samples from 820-m depth and those from 1200-m depth. We would have anticipated that in traveling 400 m through rhyolite, elements like cesium and strontium would be largely removed from solution because of their high sorption coefficients. Another difficulty in assessing our measurements at Cheshire is that we have never measured tritium concentrations as high as the value expected for the cavity region. These problems are

TABLE VI
TRITIUM CONCENTRATION IN WATER PUMPED FROM CHESHIRE

Identification Number	Date	Volume Pumped (10^6 gals.)	Tritium ($10^2 \mu\text{Ci}/\text{ml}$) ^a
C-25	10/01/85	7.4	3.82
852-1-85-008	11/04/85	9.1	3.94
C-26	11/07/85	9.3	3.97

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

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

Identification Number	Date	Volume Pumped (10^6 gals.)	Concentration		$^{85}\text{Kr}/\text{Tritium}$	
			^{85}Kr ($10^{-4} \mu\text{Ci}/\text{ml}$)	Tritium ($10^{-1} \mu\text{Ci}/\text{ml}$)	Atom Ratio ^a $\times 10^4$	Ratio ^b
852-1-83-008	09/09/83	0.06	3.0	5.77	4.5	0.76
852-1-83-009	09/09/83	0.06	3.3	5.58	5.2	0.88
852-1-84-001	08/01/84	0.17	3.2	5.38	5.2	0.88
852-1-84-003	10/23/84	3.0	2.9	5.19	4.9	0.83
852-1-84-005	10/23/84	3.0	2.4	5.14	4.1	0.70
852-1-85-001	05/09/85	3.4	3.1	4.95	5.4	0.92
852-1-85-002	05/09/85	3.4	3.0	5.00	5.2	0.88
852-1-85-003	05/14/85	3.5	3.0	4.6	5.7	0.97
852-1-85-004	05/14/85	3.5	3.2	4.7	5.9	1.00
852-1-85-005	06/18/85	4.8	2.6	4.1	5.5	0.93
852-1-85-008	11/04/85	9.1	2.4	3.9	5.4	0.92

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

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

TABLE VIII
RADIONUCLIDES IN WATER AT CHESHIRE

Radionuclide	Concentration ($\mu\text{Ci}/\text{m}\ell$) ^a	
	852-9-85-001 ^b	852-9-85-005 ^c
³ H	4.98×10^{-1}	3.94×10^{-1}
²² Na	7×10^{-9}	2×10^{-9}
⁴⁰ K	5.8×10^{-9}	9.8×10^{-9}
⁶⁰ Co	1×10^{-10}	1×10^{-10}
⁸⁵ Kr	3.0×10^{-4}	2.4×10^{-4}
⁹⁰ Sr	1.5×10^{-7}	—
¹⁰⁶ Ru	1×10^{-8}	6×10^{-7}
¹²⁵ Sb	5.1×10^{-6}	4.1×10^{-6}
¹³⁴ Cs	1×10^{-8}	9×10^{-10}
¹³⁷ Cs	2.6×10^{-6}	—
(radiochem.) ¹³⁷ Cs	1.9×10^{-6}	6.9×10^{-7}
(gamma spec.) ¹⁵² Eu	not detected	not detected
¹⁵⁴ Eu	10^{-9}	10^{-9}
¹⁵⁵ Eu	10^{-9}	10^{-9}

^aConcentrations corrected to $t_0 = 02/14/76$.

^bCollected 05/09/85; 3.4×10^6 gals. pumped.

^cCollected 11/04/85; 9.1×10^6 gals. pumped.

discussed in more detail in Ref. 9. The information collected this past year has not resolved our problems with interpreting the data from Cheshire.

IV. ANALYTICAL DEVELOPMENT

A. Measurement of ³⁶Cl (A. E. Ogard and D. J. Rokop)

The concentration of ³⁶Cl in waters pumped from RNM-2S as analyzed on the University of Rochester tandem accelerator mass spectrometer was reported in Ref. 6. We

Fig. 5. Relative tritium concentration in water from Cheshire as a function of volume pumped.

Fig. 6. Relative radionuclide concentrations in water from Cheshire as a function of volume pumped.

have recently prepared a new suite of samples that includes water taken during the previously measured period and extends the sampling period to January 1987. We prepared the ^{36}Cl samples by adding sufficient KCl to the RNM-2S water to give a $^{36}\text{Cl}/\text{Cl}$ ratio of $\sim 2 \times 10^{-12}$. We precipitated the chlorine as AgCl, purified the precipitate of any ^{36}S , and stored it in readiness for analysis sometime in early 1987.

The University of Rochester tandem accelerator mass spectrometer is the only facility in this country that can perform ^{36}Cl analyses at the level found in RNM-2S water. There are, however, circumstances in which the concentration of ^{36}Cl in the environment is higher because of anthropogenic sources and then standard mass spectrometric analysis is possible. We have prepared ^{36}Cl standards and analyzed their isotopic content with our tandem magnetic mass spectrometer to determine our lower limits of detection of this isotope. We use negative thermal ionization from a triple-filament source. The high dynamic range of measurement requires the use of a Faraday cup to collect the ^{35}Cl and single-ion counting to measure the ^{36}Cl . We obtained excellent agreement between measured and standard values for ^{36}Cl in concentration ratios of $^{36}\text{Cl}/\text{Cl}$ from 10^{-7} to 10^{-9} . The useful detection limit of our equipment as currently configured is $\sim 5 \times 10^{-10}$; the addition of an electrostatic analyzer to the tandem magnetic mass spectrometer should extend the limit to 5×10^{-11} . Although this is not sensitive enough for RNM samples, it may be sufficient for ^{36}Cl analyses of environmental samples derived from other locations.

B. Measurement of ^{99}Tc (A. E. Ogard, D. J. Rokop, D. B. Curtis, and N. C. Schroeder)

During the past year, we have continued to work on a procedure that uses the tandem magnetic mass spectrometer to measure ^{99}Tc in RNM waters. Our procedure involves adding a technetium tracer to the water sample, sorbing the technetium on an ion exchange resin, and washing it free of impurities. We then elute the technetium, remove most of the acid by evaporation, and distill the technetium to an ice-cooled surface. After dissolving the technetium in a minimum of water, we electroplate it onto a rhenium filament. We treat this filament with La_2O_3 and H_2O_2 , then with NH_4OH before introducing it into the spectrometer. The TcO_4^- ions are produced by negative ionization. The sensitivity of

this method is $\sim 5 \times 10^6$ atoms of technetium. We will apply this methodology to RNM samples during the coming year.

C. Radionuclide Transport (R. S. Rundberg, D. R. Janecky, and A. J. Mitchell)

We have made progress during the past year in understanding two aspects of radionuclide transport. The information we have acquired is only summarized here; some of it is presented in detail in another report.¹¹ We have observed that simple matrix diffusion models cannot explain our laboratory observations of radionuclide transport through fractures in natural tuff. Although nonsorbing radioactive tracers move through the fractures in reasonable correspondence with matrix diffusion model predictions, sorbing tracers produce an elution pulse earlier than expected. The size and shape of this pulse cannot be reproduced by simply reducing the distribution coefficient or adjusting the diffusivity in the model. However, by including channeling we can explain this early pulse. Channeling is caused by the irregular surface texture of the fractures and can be modeled using fractal geometry.

Another phenomenon we have been studying is anion exclusion, which results in anions moving through geologic media more rapidly than the reference water moves. We have been able to relate the magnitude of anion exclusion to the internal void volume of zeolites and clays. In the simplest terms, anions with radii larger than or close to the channel diameters of the zeolites or the interlayer spacing in the clays will be excluded from the zeolite intercrystalline pore space or the clay interlayer void. We observed good correlation between the experimentally measured anion exclusion in crushed-rock columns of tuff containing known amounts of mordenite, clinoptilolite, analcime, and smectite and the calculated interstitial and void volumes of these minerals. Anion exclusion may have a significant effect on the relative speed with which some anions travel in field situations, especially in rock that is not fully saturated with water.

D. Comparison of Los Alamos Samples with Those from LLNL (J. L. Thompson and J. D. Gallagher)

Both LLNL and LANL have reported analyses of water samples collected at essentially the same time from the same source (for example, see data in Refs. 9 and 12 about

U20n samples collected in September 1983). Direct comparison of these analyses is difficult because the laboratories chose different reference times (shot time vs collection time), but if appropriate corrections are made, it is apparent that there are significant disagreements in the reported concentrations of many radionuclides. A major source of these divergent results is the different sampling and sample treatments procedures employed by the two laboratories. These procedures are described in Ref. 8 for LANL and in Ref. 12 for LLNL. A critical step in sample treatment is the filtering done before the water is evaporated. The LLNL report presents data showing how sensitive the results are to the filtering protocol used.¹² A number of the radionuclides present in these samples appear to be associated with very small (that is, <50-nm-diam) particles that may or may not be retained on the filters—depending on the filter type and pore size. Another possible cause for differing results is the different counting techniques used by the two laboratories. To determine how important this might be, we exchanged dried samples. The sample received by LANL was identified as barrel 3, U20n, 166 ℓ, collected 10/23/83; it was accompanied by a gamma-ray analysis based on the computer code GAMANAL. We repackaged the sample to conform to the container type routinely used in our work and counted and analyzed it according to normal procedures. The computer codes we used were RAYGUN and SPECANL, which generally agree with each other within ~5%. In Table IX we have presented the LANL/LLNL activity ratios for nuclides, as determined by these two codes. It is apparent that although the counting results differ somewhat between the two laboratories, the major source of variation must reside in sample treatment before counting. We believe that part of the difference seen in the counting results can be attributed to the size difference between the LLNL sample and our usual samples. Sample size affects the source-counter geometry and, hence, the counting efficiency. This sample exchange has been helpful in that it caused us to review critically the steps in our sampling and analyzing procedures; we hope to continue and to expand interlaboratory exchanges.

V. SURVEY OF POSSIBLE SITES FOR RADIONUCLIDE MIGRATION STUDIES

Since the beginning of the RNM project, we have been aware of the need to examine the radionuclide migration in as many of the varied geological/hydrological environments

TABLE IX
SAMPLE COMPARISON

Nuclide	$\left(\frac{\text{Activity(LANL)}}{\text{Activity(LLNL)}} \right)$	
	Raygun	Specanl
⁴⁰ K	1.42	1.48
⁶⁰ Co	1.09	1.04
¹⁰⁶ Ru	1.00	1.21
¹²⁵ Sb	0.85	0.80
¹³⁴ Cs	0.93	1.01
¹³⁷ Cs	0.75	0.75
¹⁵² Eu	0.93	0.89
¹⁵⁴ Eu	1.15	1.12
¹⁵⁵ Eu	1.15	1.18

found on the NTS as possible. Our most intensively studied site, Cambric, is located in alluvial soil in which there is a low natural hydraulic gradient and little fracturing. We monitored wells drilled in the vicinity of nuclear test events in the Yucca Flat region of the NTS where there is layered tuff and where fracturing may be significant. We may have the opportunity to learn more about radionuclide movement in the Yucca Flat area in the near future (see Appendix). The brecciated rhyolites of Pahute Mesa provide yet another medium for our studies, and we are attempting to observe the movement of radionuclides in the vicinity of the Cheshire event in area 20 of the NTS. Because of the uncertainties associated with samples obtained from Cheshire (Refs. 8 and 9), we are continuing to look for other potential study sites on Pahute Mesa and have recently completed a survey of such locations.

Favorable criteria for an RNM study site include:

- (1) a test conducted below the water table so that the resulting cavity will be completely flooded with the infilling water. If appreciable amounts of postshot radioactive material are left in the unsaturated zone, it is very difficult to define the radionuclide source term because our knowledge of radioactive movement in unsaturated material is extremely limited. Much of our sampling is of water (either pumped or bailed),

and we need to know that the water is in contact with the radioactive materials of interest.

- (2) a test conducted within the past few years, preferably within less than 1 yr. A number of nuclides such as ^3H , ^{85}Kr , ^{90}Sr , and ^{137}Cs are sufficiently long-lived to be present many years after their production, but the variety of radionuclides available for sampling is much richer in the first year or two. Also, the isotopic distribution of certain shorter-lived radionuclides (for example, ^{89}Sr and ^{134}Cs) may give some clues about their initial mode of transport. Pairs of radionuclides in which one isotope has gaseous precursors and one does not (^{134}Cs and ^{137}Cs) or the isobaric precursors have differing half-lives (^{89}Sr and ^{90}Sr) are particularly useful in this regard.
- (3) a test conducted in a location for which the geologic and hydrologic features are well characterized. In particular, we would like to know the composition of the rock, the extent of fracturing, the transmissivity of the rock, and the direction and speed of water movement in the area.
- (4) a test located where there is no possibility of the presence of radioactive material from previous tests. Our understanding of radionuclide migration is too primitive to cope with the complexities associated with multiple-source terms.

Application of these criteria to tests conducted on Pahute Mesa narrows the choice of candidates for RNM studies. The majority of these tests were fired in the unsaturated zone or at the level of the water table. Only one test satisfied criteria 1 through 3. Unfortunately, it was located some 400 m from a previous test, which probably caused extensive fracturing in the area and is a potential source of radionuclides in the groundwater. It appears that Cheshire is the only site on Pahute Mesa presently available for RNM studies. We are exploring the possibility that a future test could be located in this area and thereby be suitable for our work.

ACKNOWLEDGMENTS

We greatly appreciate the efforts of C. E. Gallegos in typing this report and the editorial assistance of J. H. Heinen. We also thank M. R. Betts, M. E. Lark, and S. Lermuseaux for their assistance in gamma counting and spectral analysis.

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APPENDIX

RADIONUCLIDES AT U3kz

Early in 1984, tritium was discovered in water at an emplacement hole, U3kz, that was being readied for a test event scheduled for 1986. Subsequently, we detected the presence of fission products in water samples from this site. This discovery triggered an intensive investigation by personnel from INC, ESS, and J Divisions at LANL to discover the source and mode of travel of the observed radionuclides. This work is being funded by the weapons program and is not part of our regular RNM project. However, what we are learning from the U3kz effort is very pertinent to RNM studies and, therefore, we are including here a brief review of the U3kz work through October 1986. The review will focus largely on results obtained within the INC Division of the Laboratory.

Our initial sampling of the U3kz emplacement hole took place in April 1985 when we took two pressurized water samples. We found ^{85}Kr as well as tritium in both of them. We collected a 55-gal. sample in December 1985 during dewatering of the hole. We boiled down this barrel of water in May 1986 and detected ^{106}Ru , ^{125}Sb , ^{137}Cs , and Pu in the residue as well as on the 5- μm filters through which we filtered the water before evaporating it. Although the amount of plutonium present in the evaporated sample was quite small ($\sim 10^9$ atoms), we hoped to use the alpha activity ratio $^{238}\text{Pu}/\text{total Pu}$ to determine the source of the material. There are four expended test holes in the vicinity of U3kz that are potential sites from which radionuclides might have moved to U3kz. The alpha activity ratio $^{238}\text{Pu}/\text{total Pu}$ ranges from 0.11 to 0.24 for these four sites, so it might have been possible to associate U3kz data with one of these sites. Unfortunately, we found the U3kz alpha activity ratio value is 0.68—an anomalously high value probably caused by ^{238}Pu that was windblown and contaminated the water samples at the site.

In May 1986, we collected another 55-gal. water sample from U3kz, filtered it through 0.05- μm Millipore filters, and then evaporated it. We detected tritium and ^{90}Sr in the filtered water as well as the fission products ^{106}Ru , ^{125}Sb , and ^{137}Cs in the residue. Also, we found these same fission products in the material retained on the filters. The major fraction of the ^{106}Ru and ^{137}Cs activities occurred with the material on the filter, whereas the major fraction of the ^{125}Sb occurred with the water. In addition, we detected the

presence of ^{102m}Rh on the filters. We dissolved the filters and radiochemically separated the rhodium. Then we compared the isotopic composition of this sample with similar archived samples from the four expended test holes around U3kz. These isotopic ratios were measured with very good precision, and we have considerable confidence in them. The data are presented in Table A-I. The agreement of the $^{101g}\text{Rh}/^{102m}\text{Rh}$ values and the absence of ^{102g}Rh indicated that the rhodium detected at U3kz was produced by the Sandreef test. Although this does not prove that all the other radioactive material detected at U3kz came from Sandreef, there are no anomolous quantities of any radionuclides that would indicate contamination from other, more recent tests. Because we had on hand some archival refractory material from Sandreef, we re-counted this material with a Ge(Li) detector system and compared the results with those from the U3kz material (see Table A-II). It is apparent that many of the refractory elements present in the Sandreef cavity are not present at U3kz. The absence of ^{106}Ru in the refractory Sandreef sample is due to both its short half-life and its volatility.

The observation that radionuclides had migrated some 350 m (1150 ft) from Sandreef evoked considerable interest at the NTS as well as at LANL. The NTS site manager convened an *ad hoc* Aleman Radionuclide Migration Panel to advise him about the test shot Aleman, which was scheduled to be fired in the hole U3kz. Of particular concern was the possibility that the test shot would alter the U3kz site so that further studies could not be carried on there. This problem was resolved when LANL (J Division) drilled an auxillary hole, UE3e, 61 m (200 ft) north of U3kz to intercept the transport path of the radionuclides from Sandreef. Our analysis of water from UE3e showed that the same radionuclides were present and in the same relative amounts as in the water from U3kz. Logs taken in UE3e indicated that the measured gamma activity was confined to a narrow band at 658 m (2160 ft) depth. Sidewall coring yielded a small amount of material in which ^{137}Cs was present but not ^{106}Ru or ^{125}Sb . The reason for this apparent separation of radionuclides is not clear. Observations of the water pressure at depth in both U3kz and UE3e indicate that below ~600 m the water is under a pressure ~20 atmospheres higher than expected. We do not know the relationship between the pressurized aquifer and the radionuclide migration.

TABLE A-I
RHODIUM ANALYSES

Shot	^{102}gRh (in 1986)	$\left(\frac{^{101}\text{gRh}/^{102}\text{mRh at shot time in shot debris}}{^{101}\text{gRh}/^{102}\text{mRh at shot time in U3kz debris}} \right)$
Tortugas (1984)	measurable	1.96
Turquoise (1983)	measurable	1.03
Bouschet (1982)	measurable	1.13
Sandreef (1977)	not measurable	1.00
U3kz	not measurable	

TABLE A-II
RADIONUCLIDES PRESENT IN SAMPLES
FROM SANDREEF AND U3KZ AS OF JULY 1986 AS
DETERMINED BY GAMMA-RAY SPECTROSCOPY

	Sandreef (Refractory Soil)	U3kz (Filtered Particulates)
^{54}Mn	X ^a	O
^{60}Co	X	O
^{102}mRh	X	X
^{106}Ru	O	X
^{125}Sb	X	X
^{134}Cs	X	O
^{137}Cs	X	X
^{144}Ce	X	O
^{150}Eu	X	O
^{152}Eu	X	O
^{154}Eu	X	O

^a X = measurable; O = not measurable.

To trace the postshot movement of radionuclides from Aleman, we added tracers to the Aleman event. These consisted of a small amount of ^{244}Cm , and large amounts of NaCl, and cobalt. The ^{36}Cl and ^{60}Co activation products, along with the ^{244}Cm , would provide a variety of radiochemical species—both anionic and cationic—that would be uniquely associated with the Aleman event. In addition, we placed NaBr in UE3e before it was stemmed so that water pumped from this region could also be recognized. A number of water samples were taken from U3kz during the last dewatering before the test device was inserted. Also, surface and subsurface soil samples were collected from the area before Aleman was fired (see Fig. A-1).

Aleman was fired 9/11/86. The low yield probably resulted in incomplete activation of the tracers and little dispersion of tracer materials. We plan to drill a hole in the U3kz/UE3e area during 1987 to obtain more information concerning the geology and hydrology of the formation(s) through which radionuclide migration from Sandreef occurred. In the meantime, we are studying the particulates present in the water samples collected from U3kz during the final dewatering. Our goal is to establish if any relationships exist between the composition of the particulates, their size, and the fission products associated with them. We filtered a 55-gal. water sample through a 10- μm prefilter and an Amicon porous filter membrane; after backflushing the Amicon filter, we obtained ~ 6 g of particulates with particle diameters >5 nm. These particulates contained $\sim 10^9$ - 10^{10} atoms of ^{106}Ru , ^{125}Sb , and ^{137}Cs per gram of solid. The composition of the particulates, as determined by x-ray diffraction, was clinoptilolite (50-60%), feldspar (25%), opal (10%), and smectite ($<10\%$). We separated the clinoptilolite and smectite fractions by a grinding and settling-rate technique and found that no large differences exist in the relative concentrations of ^{106}Ru , ^{125}Sb , and ^{137}Cs associated with these two minerals. Table A-III and A-IV report results for all the samples taken by personnel from INC Division from U3kz and UE3e, respectively.

The U3kz study is continuing, and may expand to include analyses of samples from other sites in Areas 3 and 4 of the NTS.

Fig. A-1. Soil Sampling at U3kz Area .

TABLE A-III
SAMPLE COLLECTIONS U3kz

Date	Identification Number	Type	Comments
04/09/85	1234-1-85-001	water, pressure bottles	at 2100 ft
04/09/85	1234-1-85-002	water, pressure bottles	at 2100 ft
12/18/85	1234-9-85-001	water, 55 gal.	during dewatering
05/29/86	1234-9-86-001	water, 55 gal.	barrel immersed
05/30/86	1234-9-86-002	water, 55 gal.	barrel immersed
07/16/86	1234-9-86-003	water, 55 gal.	during dewatering
07/16/86	1234-9-86-004	water, 55 gal.	during dewatering
07/22/86	No. 1 thru No. 12	soil, surface	~1 in. deep, ~100 sq. in.
08/21/86	No. 1 thru No. 3	soil, subsurface	~1 ft deep in mud pit
08/21/86	1234-1-86-001	water, pressure bottles	
08/21/86	1234-1-86-002	water, pressure bottles	
08/21/86	1234-1-86-003	water, pressure bottles	
08/22/86	No. 1 thru No. 20	water, 55 gals	during dewatering

TABLE A-IV
SAMPLE COLLECTIONS UE3e

Date	Identification Number	Type	Comments
07/14/86	1235-9-86-001	water, 55 gal.	lifted in drill stem
07/14/86	1235-9-86-002	water, 55 gal.	lifted in drill stem
07/18/86	1235-9-86-003	water, 55 gal.	lifted in drill stem
07/23/86	1235-1-86-001	water, pressure bottles	at 2160 ft
07/23/86	1235-1-86-002	water, pressure bottles	at 2160 ft
07/23/86	1235-1-86-003	water, pressure bottles	at 2160 ft
07/23/86	No. 1 thru No. 3	soil, subsurface	~1 ft deep in mud pit

U.S. GEOLOGICAL SURVEY

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HYDROLOGIC ACTIVITIES OF THE U.S. GEOLOGICAL SURVEY
IN SUPPORT OF THE RADIONUCLIDE MIGRATION PROGRAM,
NEVADA TEST SITE, NYE COUNTY, NEVADA, FISCAL YEAR 1986

By William B. Scott and Charles O. Morgan

U.S. Geological Survey

ABSTRACT

Hydrologic activities conducted during the 1986 fiscal year by the U.S. Geological Survey in support of the Radionuclide Migration Program at the Nevada Test Site are summarized in this report. These activities included collecting and processing hydrologic data and entering these data into the U.S. Geological Survey, Water Resources Division, computerized ground-water data base system, managed by the Nevada State Office in Carson City.

Ground-water levels were monitored continuously at three wells and intermittently at selected wells, test holes, and emplacement holes. Selected monthly water-level measurements are listed in this report. Data collection and analysis continued on the Surface Disruption Study, and progress continued on three ground-water hydrology reports of the Nevada Test Site area.

INTRODUCTION

The U.S. Geological Survey is one of several organizations that provides services and technical expertise to the Nevada Operations Office of the U.S. Department of Energy (DOE) in support of their Radionuclide Migration (RNM) Program at the Nevada Test Site (NTS). The primary goal of the RNM Program is to provide the necessary data and the interpretation of these data to demonstrate to the scientific and public communities, whether past, present, and future activities at the NTS will or will not adversely affect the quality of ground water in the area.

The purpose of this report is to summarize the hydrologic activities of the Geological Survey, during the 1986 fiscal year, in support of the DOE-RNM Program at the NTS. The report provides the following: (1) tables of selected water-level measurements for three wells monitored continuously by means of automatic measuring equipment; (2) tables of water-levels measured intermittently at selected wells, test holes, and emplacement holes; and (3) a description of work done on projects and reports related to the RNM Program. This report is written specifically as part of the DOE 1986 Annual Radionuclide Migration Program Summary Report.

HYDROLOGIC ACTIVITIES

The U.S. Geological Survey, NTS-Weapons Testing Hydrology Program staff, located in Las Vegas, Nev., provides hydrologic and hydrogeologic technical expertise to the DOE-RNM Program. This is done by collecting, processing, and analyzing hydrologic and hydrogeologic data; planning and conducting hydrologic investigations; serving on the RNM Committee; and producing technical publications of interpretative results. The following sections of this report summarize the Geological Survey's activities in support of the RNM Program for the 1986 fiscal year.

Monitoring of Ground-Water Levels

Continuous Measurements

Ground-water levels were monitored continuously, by means of automatic measuring equipment, at the Nash Satellite well (UE-2ce), Bourbon Satellite well (UE-7nS), and the Chesire well (U-20n PS#1DD-H). In January 1986, each well was equipped with (1) a calibrated pressure transducer for measuring the water level, (2) a micrologger with a microchip data-storage module for continuous recording of data received from the transducer, and (3) solar panels with storage cells to provide power to operate the transducer and micrologger. The data stored in the microchip data-storage module were converted to water-level measurements, in feet with reference to land-surface datum, which is a datum plane that is approximately at land surface at each well. These measurements will be reviewed, then stored in the Ground-Water Site Inventory system, the U.S. Geological Survey computerized data-base system for storage and retrieval of ground-water information. Selected monthly water-level measurements for the Nash and Bourbon Satellite wells, and the Chesire well are shown in tables 1, 2, and 3, respectively. A comparison of changes in water levels measured at these wells is described below.

Nash Satellite Well--Area 2

On September 8, 1977, the water level measured in the Nash Satellite well (UE-2ce) was 1,407.17 feet below land-surface datum. A comparison of this water-level measurement with the September 18, 1986, measurement in table 1 indicates that the water level declined 72.98 feet during this 9-year period. Data in table 1 also show a water-level decline of 11.52 feet during the period from April 15, 1986, to September 18, 1986.

TABLE 1.--Selected water-level
measurements in well UE-2ce

[Depth, in feet below land-
surface datum]

Date		Water Level
Jan.	1986	*
Feb.	1986	*
Mar.	1986	*
Apr.	15, 1986	1,468.63
May	15, 1986	1,473.93
June	15, 1986	1,476.53
July	15, 1986	1,478.07
Aug.	13, 1986	1,479.27
Sept.	18, 1986	1,480.15

* Transducer malfunctioning.

Bourbon Satellite Well--Area 7

On September 8, 1977, the water level measured in the Bourbon Satellite well (UE-7nS) was 1,968.12 feet below land-surface datum. A comparison of this water-level measurement with the September 14, 1986, measurement in table 2, indicates that the water-level declined 10.30 feet during this 9-year period. Data in table 2 show that the water level in this well has been relatively stable from January 1986 through September 1986, except during periods when the well was being pumped.

TABLE 2.--Selected water-level
measurements in well UE-7nS

[Depth, in feet below land-
surface datum]

Date	Water Level
Jan. 16, 1986	1,978.02
Feb. 15, 1986	1,977.88
Mar. 04, 1986	1,977.75
Apr. 15, 1986	1,995.88*
May 14, 1986	2,000.48*
June 15, 1986	2,014.42*
July 15, 1986	1,983.16
Aug. 15, 1986	1,978.83
Sept. 14, 1986	1,978.42

* Well being pumped.

Cheshire Well--Area 20

The water level in the Cheshire well (U-20n PS#1DD-H) was monitored continuously from June 1986 to August 1986, then measurements were made intermittently with an electric cable well-measuring device commonly called the "Iron Horse." This device is designed especially for measuring water levels in deep wells. The continuous water-level monitoring equipment had to be removed in August 1986 because the pump equipment was removed from the well. During the period of continuous monitoring, the water levels recorded showed a daily fluctuation of 5 feet. This unusual fluctuation in water level is being re-examined and is considered to have been associated with the electrical equipment. On April 15, 1981, the water level in well U-20n PS#1DD-H was reported to be 2,035 feet below land-surface datum. A comparison of this measurement with data in table 3 shows that the water level in this well may have been relatively stable since 1981.

TABLE 3.--Selected water-level measurements
in well U-20n PS#1DD-H

[Depth, in feet below land-surface datum]

Date	Water level
Jan. - Apr. 1986	*
May 15, 1986	2035.12
June 15, 1986	2036.53
July 15, 1986	2036.18
Aug. 11, 1986	2037.06
Sept. 1986	*

* Unable to measure well.

Intermittent Measurements

The water level in selected wells and test holes was measured intermittently with the Iron Horse. A hoist unit with sampling barrels was used to obtain water samples from the wells and test holes for radiochemical analysis.

Faultless Well

The Faultless well (UC-1 P2sr), located in Hot Creek Valley, central Nevada, is the only site in the RNM Program not on the NTS. The water level in this well was measured and a water sample was collected in July 1986. The water level was 960 feet below land-surface datum. This is a rise in water level of 33.6 feet since October 1985, which is 395.2 feet below the estimated preshot-water level. The water temperature in the well ranged from 66.2 °F at 990 feet below land-surface datum to 78.8 °F at 2,590 feet below land-surface datum. Radiochemical analysis of the water sample was done by Reynolds Electrical & Engineering Co.¹ laboratory located at the NTS.

¹ The use of firm names in this report is for identification purposes only and does not constitute endorsement by the U.S. Geological Survey.

Selected Wells and Test Holes

Water levels were measured and water samples collected intermittently from selected wells and test holes at the NTS. Some emplacement holes were measured and sampled intermittently prior to the conducting of weapons tests. Water-level measurements are critical inputs to the interpretation of the ground-water hydrology of the NTS, especially in areas where the direction of ground-water movement has not been clearly defined. The results obtained from water-sample analyses, in conjunction with water-level measurements are necessary for detecting radionuclide migration and defining the direction and rate of movement. Water levels measured in selected wells and test holes are shown in table 4. Table 5 shows water levels measured in selected emplacement holes.

TABLE 4.--Water levels measured in selected wells and test holes

[Depth, in feet below land-surface datum]

Site identification	Date	Water level
Pahute Mesa		
Ex. Hole #2	May 1, 1986	851.50
UE-14 b	May 28, 1986	1,666.04
Test Well B	June 10, 1986	1,504.23
UE-18 r	June 12, 1986	1,467.86
UE-18 t	June 12, 1986	1,366.24
UE-3 e	June 27, 1986	1,555.72
Test Hole 7	June 30, 1986	1,604.02
Test Well D	June 30, 1986	1,722.79
UE-17 a	July 8, 1986	638.97
UE-1 l	July 8, 1986	517.89
UE-1 a	July 8, 1986	545.02
UE-1 c	July 8, 1986	1,296.49
UE-1 b	July 10, 1986	644.26
UE-6 e	July 10, 1986	1,509.46
UE-6 d	July 10, 1986	1,516.05
UE-5 n	July 10, 1986	703.99
UE-16 f	July 14, 1986	367.54
UE-10 ITS #1	July 14, 1986	1,221.37

TABLE 5.--Water levels measured in selected
emplacement holes

[Depth, in feet below land-surface datum]

Site identification	Date	Water level
U-3kv	May 12, 1986	1,534.58
	May 27, 1986	1,533.42
	June 9, 1986	1,533.98
	June 22, 1986	1,533.56
U-7ca	May 27, 1986	1,702.75
	June 9, 1986	1,703.91
	June 16, 1986	1,703.08
	June 22, 1986	1,703.08
U-7cb	Aug. 4, 1986	1,789.07
	Aug. 7, 1986	1,789.27
U-19an	Dec. 26, 1985	2,106.65
	May 13, 1986	2,113.41
U-20ap	Dec. 26, 1985	2,138.22
U-20aq	Dec. 26, 1985	1,882.99
U-20as	Apr. 22, 1986	2,012.12
	Apr. 23, 1986	2,012.94
	Apr. 25, 1986	2,012.88
	Apr. 28, 1986	2,013.07
U-20av	June 6, 1986	2,013.04
	Aug. 4, 1986	2,080.01
	Aug. 5, 1986	2,080.14
	Aug. 7, 1986	2,079.88

Surface Disruption Study--Area 3

The U.S. Geological Survey is conducting a study of surface disruption features, craters, formed by ground-surface subsidence associated with underground nuclear testing at the NTS. These craters have created the potential for ground-water recharge to occur in areas that normally produce relatively small quantities of recharge. Increased recharge by means of the craters increases the potential for radionuclides to be transported from the underground detonation chamber into the regional ground-water flow system. The primary objective of the study is to analyze the water budget at selected craters and obtain quantitative measurements of recharge. Two craters in Area 3, referred to as craters 1 and 3, were selected for this study because of their proximity to each other and their ability to be representative of most craters in the area.

The approach is to determine the significance of the craters as a source of ground-water recharge by obtaining bare-soil evaporation, soil-moisture deficit, and meteorological data. These data will be used as input to a water-balance model. Comparison of model results for each site should aid in determining the significance of the craters as a source of ground-water recharge. If increased ground-water recharge is occurring as a result of the craters, estimates can be quantified.

Field-data measurements begun in April 1985 were discontinued in June 1986. These measurements included the collection of (1) meteorological data, (2) precipitation data, (3) parameters for determining bare soil evaporation, and (4) periodic measurement of soil moisture.

The preliminary processing of field data was completed for the meteorological stations, precipitation measurements, Idso stations (named for the Idso-Jackson equation used in data analyses), and soil-moisture logs. Evaluation and analysis of the large volume of data collected continued. Preliminary interpretation of the data is in progress.

Computerized Ground-Water Data Base

In February 1986, the U.S. Geological Survey State Office in Carson City, Nev., established a computerized data base for the storage of current and historical ground-water information for Nevada. Data for approximately 300 wells and test holes at the NTS were entered into this data base during FY 1986. Data for wells in parts of Nevada surrounding the NTS are being stored also and can be used in the interpretation of ground-water movement at the NTS.

Numerous volumes of hydrologic-data files accumulated over the past 30 years for the NTS were transported from the U.S. Geological Survey Nuclear Hydrology Office in Denver, Colo., to the Weapons Testing Hydrology Program Office in Las Vegas, Nev. Organization of these data files according to areas and sites, was started and will serve as the main source of information to be entered into the computerized data base. These data will be used to gain a better understanding of ground-water flow patterns and the direction and rate of radionuclide migration at the NTS.

Technical Support

The Geological Survey continued to provide technical support to the DOE- RNM project by attending each of the quarterly RNM Committee meetings and by assisting in the development and review of the drilling plan and proposal for the U-20n downgradient test hole.

The purpose of the RNM Committee, which consists of representatives from U.S. Department of Energy, U.S. Geological Survey, Desert Research Institute, Los Alamos National Laboratory, and Lawrence Livermore National Laboratory, is to advise, direct, and conduct research associated with radionuclide migration. The Geological Survey provided technical expertise in hydrology and ground-water hydraulics for the planning, conducting, interpreting, and reviewing of DOE-funded RNM projects.

The Geological Survey provided technical assistance in the development of plans and procedures for the drilling, testing, and sampling of the U-20n downgradient test hole on Pahute Mesa. The purpose of the test hole is to determine if radionuclides are migrating upward from the Chesire cavity and then moving laterally through an upper permeable formation. Technical assistance was provided by (1) assisting in the review and rewriting of the drilling specifications, (2) reviewing existing technical reports and data concerning the Pahute Mesa area to determine the distribution of vertical hydraulic heads and identifying potentially permeable zones in the area, and (3) providing predictive drawdown data by using digital simulation to solve analytical equations for various scenarios of pumping from the U-20n downgradient test hole.

REPORTS IN PROGRESS

Work continued on the following three ground-water related reports being prepared by the U.S. Geological Survey:

1. The NTS Hydrologic Atlas--hydrogeologic data collected from selected wells and test holes in the NTS area are presented in tabular format.
 2. The Pahute Mesa Ground-Water Map--a map showing the ground-water altitude contours for the Pahute Mesa area.
 3. The Yucca Flat Ground-Water Map--a map showing the ground-water altitude contours for the Yucca Flat area.
- These publications contain hydrologic data needed to determine the direction and rate of ground-water flow, which will be used in evaluating radionuclide migration at the NTS.

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