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The Cambric Migration Experiment

A Summary Report

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The Cambric Migration Experiment

A Summary Report

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THE CAMBRIC MIGRATION EXPERIMENT

A SUMMARY REPORT

by
Ernest A. Bryant

ABSTRACT

The Cambric migration experiment was undertaken in 1974 to provide data concerning the migration of radionuclides away from the site of a nuclear test where there is an induced hydraulic gradient. First, the distribution of radioactive isotopes around the site of the Cambric explosion was investigated by analysis of samples recovered from a hole drilled through the nuclear debris. Then water was pumped from a well, located 91 m away, to induce flow from the nuclear explosion site. Analyses of water samples showed that the migration velocity of tritium, ^{36}Cl, ^{85}Kr, ^{99}Tc, ^{106}Ru, and ^{129}I was nearly the same as that of the moving water, from the explosion site to the pumped well. Less than 0.5% of the total ^{90}Sr and 0.0003% of the total ^{137}Cs accompanied the tritium to the pumped well, although both isotopes appear to have migrated away from the source zone to some extent. The concentration of ^{239}Pu at the pumped well was below the detection limit of 10^6 atoms/mℓ in water collected at the time of peak tritium concentration. Peak tritium concentration in the pumped water occurred when 5,000,000 m^3 had been pumped.

Now that the elution of the mobile radionuclides is nearly complete, it is time to review and revise plans for future measurements at the Cambric site.

I. INTRODUCTION

A committee of scientists is regularly convened by the Nevada Operations Office of the Department of Energy (DOE/NVO) to review progress for the Hydrology and Radionuclide Migration Program (HRMP) and to make recommendations for future

directions. The steering committee has urged that "... a comprehensive report on activities at the Cambric site since the Hoffman, Stone, and Dudley report ..." be written. The present report has been compiled in response to the steering committee recommendation.

Nearly all of the experiments discussed here have been reported previously elsewhere, and many individuals have contributed to the results reviewed and discussed here. The author has attempted to be accurate in attributing credit, but there are undoubtedly errors of omission, or otherwise, for which he apologizes. The references cited represent the sources for the material presented and are not intended to be a comprehensive bibliography of Cambric-related publications. When appropriate, the author's comments are printed in italics to distinguish them from comments taken from the many reports and papers that were the source of information in this report.

II. NUCLEAR TESTING

The Nevada Test Site (NTS), located about 60 miles northwest of Las Vegas, has been used for contained underground tests of nuclear explosive devices since 1963. In preparation for a test, a hole is drilled to a depth sufficient to prevent the venting of radioactive debris to the atmosphere and the device is placed at the bottom of the hole. After the device is placed in the hole, the hole is plugged and the test device is exploded. In some cases, the hole penetrates to below the level of the water table and then, after the test, water flows back in to surround and infiltrate the debris left by the explosion.

An underground nuclear explosion almost instantly creates a cavity filled with vaporized material at high pressure and surrounded by a layer of molten rock. As the gases cool and condense, the molten rock flows to the bottom of the cavity. At some time the cavity pressure drops sufficiently to allow the overburden to cave in, beginning the formation of a chimney. This chimney formation process may continue all the way to the surface, where it results in a crater, or it may stop at some intermediate point.

Each nuclear device contains radioactive materials, and the nuclear reactions involved in the explosion produce more radioactive materials. Thus a substantial inventory of radioactive materials has been emplaced in the ground at the NTS as a result of nearly 30 years of underground nuclear testing. From the beginning, the Atomic Energy Commission, then the Energy Research and Development Administration (ERDA) and now the DOE, have recognized the need for knowledge about both the potential for and the rate of groundwater transport of test-related radioactive materials.

A substantial fraction of the radioactive debris from underground nuclear explosions is contained in the glass formed after explosive melting of rock surrounding the tested device. However, some fraction of the debris, especially volatile elements such as tritium and the noble gases, is not contained in such glass. Based on observations made during numerous drillback operations, it is clear that some of the more volatile radioactive materials, including gases, find their way into the chimney rubble, probably during chimney formation. Furthermore, the glass itself is not a permanent container but is expected to both weather (leach) and devitrify over time; there is also a possibility for

concurrent release of contained radionuclides into the groundwater. Information relevant to the migration of radionuclides at the NTS was reviewed in 1976 by Borg et al.[1]

III. THE HYDROLOGY AND RADIONUCLIDE MIGRATION PROGRAM

The HRMP (formerly the Radionuclide Migration program), sponsored by the DOE/NVO, has gathered information about the distribution of test-produced radioactive materials and about the local and regional hydrology as it relates to migration (groundwater transport) at the NTS. Such information is needed to assess whether the underground nuclear tests have created, or are likely to create, a threat to the public or the environment through the deposition of radioactive materials that might then be transported by groundwater to some offsite location.

IV. THE CAMBRIC EXPERIMENT

In 1974, concern about possible long-term environmental consequences of the underground testing of nuclear devices at the NTS led the ERDA to propose a study of the groundwater transport of radioactive materials away from the site of a small nuclear explosion called Cambric. This 0.75-kt test was conducted on May 14, 1965, at a depth of 294 m in alluvium at Frenchman flat at the NTS. Groundwater had already flowed into and filled the explosion cavity region by 1974, when the transport study began.

The idea was to first measure the distribution of radioactive materials in water and rock in the vicinity of the explosion. The second step was to induce an artificial hydraulic gradient by pumping water from a nearby well and to regularly test the pumped water for the presence of radioactive species that might have migrated from the explosion cavity.

Thus the first RNM project, the Cambric migration experiment, was a unique test of the transport of radionuclides by groundwater flowing along an artificially induced hydraulic gradient from the Cambric debris to a nearby pumped well. Before the pumping began, an extensive study was conducted to determine the form and distribution of radioactive materials at and around the Cambric detonation point; that is, the "source term." The results of that study have been thoroughly documented[2,3,4,5]; some of the highlights are included here for completeness and the convenience of the reader.

V. THE GEOLOGIC SETTING

An extensive description of the geologic-hydrologic setting for the Cambric experiment appears in the report by Hoffman, Stone, and Dudley.[2] This information is supplemented by a review and analysis of the hydrology by Wheeler, Ng, and Logan,[6] and also by Burbey and Wheatcraft.[7] The geology and hydrology, briefly summarized here, is based on information appearing in these reports.

Frenchman Flat, the site of the Cambric explosion, is located in the southeastern part of the NTS, about 100 km northwest of Las Vegas, Nevada. The flat is a closed basin with drainage to the normally dry Frenchman Lake at an altitude of

938 m. Gently sloping desert of sparsely vegetated alluvial soil surrounds the playa up to about 1030 m altitude. Mountainous terrain up to 1900 m surrounds most of the basin.

Well-defined drainage channels in the mountainous areas give way to braided and slowly migrating channels on the middle and lower desert floor. These ephemeral channels carry runoff only during major storms and lose it rapidly as a result of infiltration and evaporation; they only rarely supply water to the central playa.

The paleozoic basement carbonate rocks range in age from late Cambrian through Devonian. About 25 Myr ago, volcanic eruptions covered the area with thousands of meters of tuffs and lava flows. Tectonic activity subsequently created a series of north-trending mountain ranges. Erosion of the ranges created the tuffaceous alluvium that makes up the upper stratum (~500 to 800 m) of the Frenchman Flat basin.

The alluvium at the Cambric site is dominantly medium to coarse sand; there are pebbles in most samples and gravel and cobble in the upper 200 m. The lithologies were mainly derived from the Tertiary volcanics and include only occasional Paleozoic quartzite, dolomite, and limestone. Zones with finer grain sizes are accompanied by zeolitization and argillization of the sand-silt matrix. A sample of bluish-grey silty clay came from a depth of 292 m at the Cambric site.

Frenchman Flat lies within the Ash Meadows groundwater system, which flows in a generally southwesterly direction. The principal aquifer is located in the underlying carbonate rocks. Measured gradients are about 0.01 vertically and 0.001 in a southeastward direction. The flow, if any, at the Cambric site itself may be either downward to the carbonate aquifer or horizontal. It is thought that there are aquitards in which the conductivity is an order of magnitude lower than that of the alluvium surrounding the Cambric explosion site. There apparently is insufficient information to come to a firm conclusion about the flow direction or velocity. It is generally agreed that the flow is small, but Wheeler asserts that it might be significant while Wheatcraft concludes it is probably insignificant relative to the Cambric migration experiment. Further information about the hydrologic parameters is contained in the section on hydrologic modeling.

VI. THE SOURCE TERM

In preparation for the migration experiment, several wells were drilled at the Cambric site. In May of 1974, well RNM-1 was drilled through the deposit of radioactive debris left by the Cambric explosion. The first 146 m of this well followed the original slant-drilled, diagnostic sampling hole, but a new hole was drilled below that point.

An extensive series of water and core samples were collected from this hole and analyzed. Because many of the shorter lived radioactive isotopes had decayed away in the 9 yr between the nuclear test and the analyses, only a limited number of radionuclides were present in sufficient concentration for analysis.

The general observations listed below were based on results from analyses of the core samples.

- The tritium content of water recovered from core samples was highest for cores from the cavity area. Almost all the tritium was present as tritiated water (HTO). A small fraction, less than ~0.1% of the total tritium, was present as tritiated hydrogen or methane (HT or CH_3T).

- The ratio of ^{85}Kr to tritium in core water varied from 0.4 to 5 times the expected ratio for the device as a whole, suggesting that a substantial fraction of the ^{85}Kr could be found in water.

- With the exception of those taken in the old diagnostic hole, cores from above the water table exhibited background levels for tritium, ^{85}Kr, and other radionuclides. The old hole was probably contaminated during the diagnostic sampling process in 1965.

- The radioactive debris was concentrated in fused, glassy material in the vicinity of the explosion cavity. The ^{147}Pm, ^{155}Eu, ^{241}Am, and ^{239}Pu were all highly concentrated in the fused debris. This was attributed to their refractory nature; it is thought that they condensed from the vapor before the chimney collapse began. ***It seems likely that the debris contained significant concentrations of gaseous fission products such as ^{85}Kr in addition to most of the refractory products.***

- Relative to ^{239}Pu and ^{155}Eu, the volatile species (^{125}Sb) and the fission products with gaseous or volatile precursors (^{90}Sr, ^{137}Cs, and to a lesser degree ^{144}Ce) were depleted in the fused materials from the cavity region. They were sometimes associated with nonfused material and also tended to be relatively more concentrated in cores from the chimney.

- Analysis of water extracted from two solid-core samples indicated that ^{90}Sr, ^{137}Cs, and ^{239}Pu were more concentrated in the solid than in the water by factors (effective distribution coefficients) of $\sim 10^4$, $>10^4$, and $\sim 10^8$, respectively.

Analyses of water samples pumped from five isolated zones* of RNM-1 provided information about the initial distribution of tritium and other radionuclides in the groundwater below, in, and above the cavity region. By comparing the measured radionuclide-to-tritium ratios with the ratio calculated for the Cambric source, the authors derived depletion factors that were "... regarded as the retention factor for the particular radionuclide on the solid debris." Several observations were reported.

- No radioactive species could be detected in water pumped from a zone 50 m below the bottom of the cavity.

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* The five zones were Zone I: below the cavity (~359 m vertical depth); Zone II: lower cavity (~302 m); Zone III: upper cavity (~290 m); Zone IV: chimney area (~277 m); and Zone V: periphery of the chimney area (~263 m).

- The concentration of tritium in the cavity region, at t_0 + 10 yr,* was 8 - 14 x 10^6 dpm/mℓ, or ~2000 times greater than the concentration guide for an uncontrolled area. The concentration of ^{239}Pu was more than 600 times lower than the guide. Typical concentrations of several radionuclides for five regions are listed in Table I, which is adapted from the Hoffman, Stone, and Dudley report.[2]

- Burbey and Wheatcraft[7] derived a weighted average tritium concentration of 6.53 μCi/mℓ (at t_0) for the cavity area; this corresponds to 8.3 x 10^6 dpm/mℓ at t_0 + 10 yr.

- The tritium concentration in the chimney was ~2% of that in the cavity. If the cavity contained the same total tritiated water volume as the chimney, then ~98% of the total tritium was in the cavity. There is no information about the actual volumes, but Hoffman, Stone, and Dudley used the concentration in the cavity region and a nominal value for porosity to estimate that a "T-exchange radius" of ~16 m was required to account for the total tritium present. Burbey and Wheatcraft assumed an 18-m radius for their calculations. *A tritiated sphere with this radius centered on the Cambric explosion should have been sampled at Zone IV (the chimney area).*

- The highest retention factors (>10^6) were observed for ^{239}Pu and ^{147}Pm. These high values were attributed to the formation of insoluble hydroxides.[2] It was also noted, however, that a large fraction of the ^{239}Pu, and of other refractory species as well, is encapsulated in glass and is presumably unavailable for immediate dissolution.

- Retention factors of ~10^2 were reported for ^{90}Sr, ^{106}Ru, and ^{125}Sb; ~10^4 for ^{137}Cs; and ~10^3 for ^{144}Ce.

- The measured ratio of ^{85}Kr to tritium relative to that expected for the Cambric source ranged from a low of 0.3 in the cavity to a high of 1.5 in the chimney for representative pressurized water samples. The concentration in the cavity was ~10 times higher than in the chimney.

VII. THE MIGRATION EXPERIMENT

After the source term had been defined, the migration experiment began. Because the velocity of groundwater at the Cambric site is quite low,[2] it was necessary to produce an artificial gradient so that water would move to the nearby well in a reasonable length of time.

Well RNM-2S had been drilled vertically to a depth of 350 m at a point 91 m south of the center of the Cambric detonation point. RNM-2S was drilled before the drilling and sampling of RNM-1 to minimize the possibility for contamination. The portion of the well below the water table was gravel-packed and the casing

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* In this report, data for the level of radioactivity have generally been corrected to the Cambric explosion time, t_0, on May 14, 1965; exceptions are noted.

Table I. Representative Activity Levels in Pumped Water Samples

Water Source	T	^{85}Kr	^{90}Sr	^{106}Ru	^{125}Sb	^{137}Cs	^{239}Pu
	(dpm/mℓ at t_0 + 10 yr)						
Below Cavity	No radioactivity above background detected						
Low Cavity[a]	7.56×10^6		8.8	8.3	12.0	1.98	0.014
Low Cavityb[c]	1.36×10^7	347	7.9	10.9	4.6	1.58	0.0029
High Cavity	8.52×10^6	516	5.3	3.8	2.3	1.35	0.0026
Chimney	1.84×10^5	37	4.6	nd[c]	nd	0.79	nd
Periphery	6.2×10^4	6	0.2	nd	nd	0.16	nd
Concentration Guide[d]	6.7×10^3	--	0.67	22.2	222	44.4	11.1

[a] After pumping 1.9 m^3.
[b] After pumping 11.5 m^3.
[c] nd = not detected.
[d] This was the recommended concentration guide applicable to water in uncontrolled areas in 1976.

was perforated at various depths from 325 to 350 m; thus, the intake zone for RNM-2S was located 16 to 41 m below the bottom of the Cambric cavity.[6] A pump was installed and pumping began in October 1975. Pumping was nearly continuous, first at 300 gpm and then at 600 gpm, until the pump was "permanently" shut down on August 29, 1991. The history of pumping from 1975 to early 1981 is shown in Table II, which was adapted from Ref. 7. From 1981 to 1990, the pump was occasionally shut off for a few days. The pump was also shut down from September 25, 1990 until December 3, 1990, and from December 18, 1990, until February 4, 1991.

Well RNM-1 was reconfigured for sampling during the migration experiment by leaving the pump and packers positioned to collect water from the chimney and adjacent peripheral area (Zones IV and V). Based on separate tests of these zones, it was anticipated that most of the water would come from the chimney in this configuration; therefore, subsequent references to water pumped from RNM-1 with this configuration refer to the "chimney" as the source.

VIII. SAMPLING

The water being pumped from well RNM-2S was sampled on a regular basis. Water samples were collected less frequently from the chimney area in well RNM-1. The samples from both wells were analyzed for tritium and other radioactive species.[8-17] Results of these analyses are summarized and discussed in the following sections.

Table II. Pumping History at Well RNM-2S

Date Off	Date On	Days Off	Day in Sequence of Pumping
3/7/80	3/24/80	17	1614-1631
6/6/80	6/16/80	10	1706-1716
11/10/80	11/19/80	9	1864-1873
1/13/81	2/10/81	28	1929-1957

IX. TRITIUM

Tritium should be a nearly ideal tracer for groundwater in the porous alluvium surrounding the Cambric site. Just what constitutes ideal tracer behavior is, of course, a matter of choice. For our purposes, an ideal tracer would be a non-reactive species that moves with and is not excluded from any region containing mobile water.

Groundwater may be considered to consist of a mobile fraction and an immobile fraction. In the presence of a hydraulic gradient, there is a wide distribution of instantaneous velocities within the mobile fraction. The immobile fraction consists of water trapped in the solid matrix; for example, in crystals. To the extent that trapped water can exchange with mobile water, it might also be considered mobile, but such water would not retain any dissolved species when it entered into the trapped state and our interest is focused on the transport of dissolved, or suspended, species.

Any deviation of the average tritium velocity from the mobile water velocity would require sorption of HTO, or the exchange of T^+ (or OT^-) with H^+ (or OH^-), at mineral surfaces. In a system where the ratio of mobile water to trapped water is large, such deviations should be small; with a porosity in the 30% range, the alluvium surrounding Cambric might be expected to have only a small fraction of trapped water. Thus deviations of the tritium velocity from the water velocity are expected to be relatively small, but may not be zero.

Tritium was, by far, the most abundant radioactive species in water from the cavity region at Cambric. For this reason and also because it is a readily analyzed tracer for the Cambric water, water samples collected from the pump at RNM-2S or from RNM-1 were routinely analyzed for tritium. Results of these analyses are shown in Fig. 1, where tritium concentration is plotted against total volume pumped from well RNM-2S. One can note four features of the two curves.

- The drop in tritium concentration in the chimney zone at well RNM-1 was initially precipitous but soon slowed to an exponential decay with a rate of 0.5×10^{-6} m^{-3}. The rate during the initial rapid drop was $\sim 3.5 \times 10^{-6}$ m^{-3}.

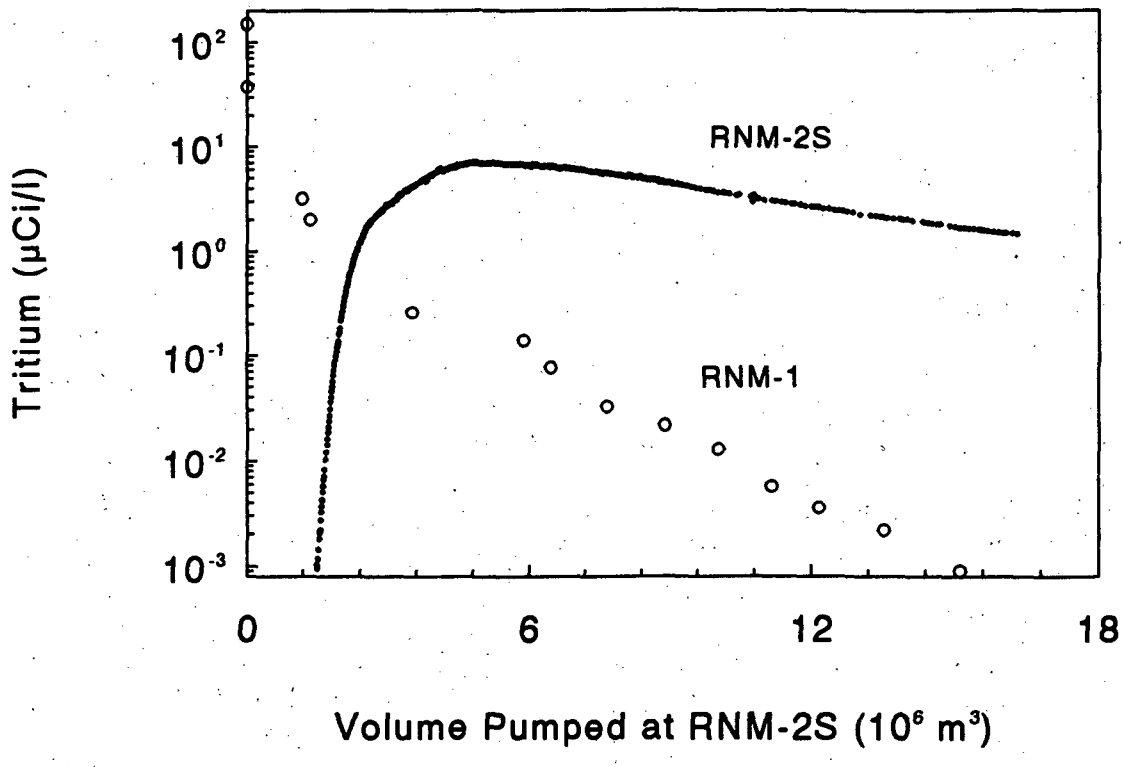

Fig. 1. Tritium concentration at RNM-1 and RNM-2S *vs* volume pumped at RNM-2S.

- The first measured tritium concentrations above background appeared in RNM-2S water at a pumped volume of ~1.3 Mm^3.*
- The rise in tritium concentration at RNM-2S was also very rapid, increasing exponentially at an initial rate of ~12 x 10^{-6} m^{-3}; the decay after the peak was much slower at a rate of 0.16 x 10^{-6} m^{-3}.
- The peak concentration of tritium at RNM-2S was ~7 nCi/mℓ (at t_0) and occurred at a pumped volume of 4.9 Mm^3. This concentration is a factor of 10^3 smaller than the 6 to 11 μCi/mℓ reported to be representative in the cavity region at Cambric (see Table I).

The tritium data were compared to hydrologic model calculations by Travis and Rundberg,[11] Wheeler,[6] and Burbey and Wheatcraft.[7] A discussion of the models and comparisons can be found below in the section on hydrologic modeling.

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* "Mm^3" is used here as an abbreviation for million cubic meters.

X. KRYPTON (^{85}Kr)

The noble gases from helium to xenon are often found dissolved in groundwater; for example, ^{4}He accumulates in groundwater as a result of alpha-particle decay and its concentration has been used to calculate the age of groundwater. Thus it was expected that noble gases from the Cambric source would be found in and transported by groundwater.

The fission product ^{85}Kr, with a 10-yr half-life, is formed in part directly by fission and in part by decay of its short-lived bromine, selenium, and germanium fission-product precursors. Only a fraction of the ^{85}Kr and its relatively volatile precursors is condensed into the molten glass. Thus, once the precursors have decayed, only a fraction of the total ^{85}Kr is trapped in the refractory solid debris.

Numerous ^{85}Kr analyses have been carried out during the RNM Cambric project. From the beginning, the results for ^{85}Kr and its ratio to tritium have been disturbingly variable. Analytical results for sequential samples from the same source have sometimes differed by as much as a factor of 2.

The source of the variability has not been identified.[8] *It is difficult to imagine a process that could produce and retain the observed variability in the groundwater before it was pumped or sampled. It is less difficult to imagine a process taking place during sampling that could result in the collection of variable samples and one can imagine that there might be uncontrolled variations in the analytical procedure itself. For example, if small gas bubbles were to form on surfaces near the pump inlet as a result of the local decrease in pressure and then subsequently become detached, one would expect to observe random variations in the apparent ^{85}Kr concentration reflecting fluctuations in the rate of bubble detachment.*

Based on the observed $^{85}Kr/T$ ratios for RNM-1 water and core samples, Hoffman, Stone, and Dudley[2] concluded that the data were ".. consistent with the calculated ratio for Cambric...." In spite of the wide range of ratios, which varied from 0.18 to 3.0 times the expected value, it does seem clear that a substantial fraction of the ^{85}Kr from the Cambric explosion was present in the water around the detonation site when the migration experiment began.

The data indicate that there were variations in the $^{85}Kr/T$ ratio for water in different zones in the cavity-chimney area; such fractionation could easily result from processes taking place during the time required for decay of the ^{85}Kr precursors following the explosion. Relative to tritium, the ^{85}Kr appears to have been concentrated in the chimney and depleted in the cavity. The $^{85}Kr/T$ ratio was a factor of 3 or 5 larger in the chimney than in the cavity. However, the tritium concentration in the chimney was only ~2% of that in the cavity, suggesting that most of the dissolved ^{85}Kr, as well as the tritium, was in the cavity zone.

The observations below were made concerning the data from numerous RNM-1 and RNM-2S samples. These data are shown in Fig. 2 as ^{85}Kr concentration *vs* volume pumped from RNM-2S.

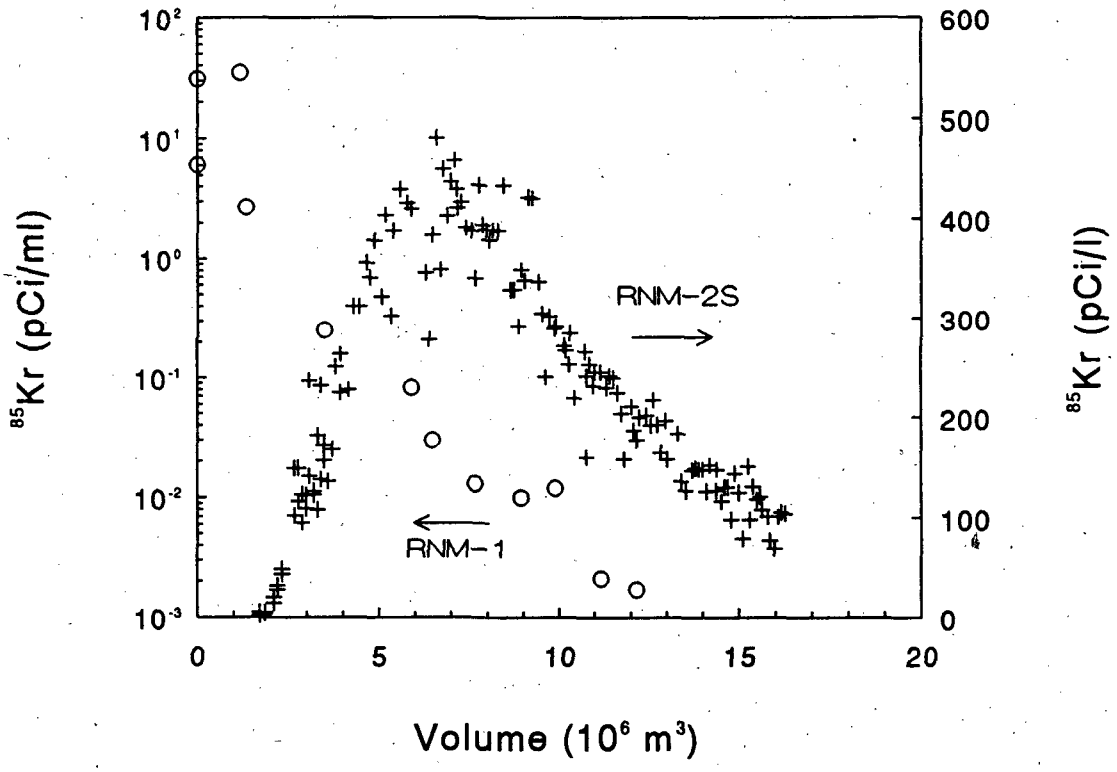

Fig. 2. ^{85}Kr concentration *vs* volume pumped from well RNM-2S.

- The ^{85}Kr concentration has dropped in the chimney zone at well RNM-1, but the initial drop was not quite as precipitous as the initial drop in tritium concentration. Beginning at a pumped volume of 6 Mm^3, the ^{85}Kr concentration in RNM-1 water decreased at the same rate as the tritium concentration.

- The ^{85}Kr/T ratios were high in the chimney zone in 1975 (~1.5 times the expected ratio) and increased to 75 times the expected ratio when pumping began. Subsequently, the ratio dropped to ~3 times the expected. This suggests that the ^{85}Kr was initially distributed rather differently than the tritium in the chimney area, or became differently distributed during the 10 yr between the explosion and the beginning of the migration experiment.

- At RNM-2S, the ^{85}Kr was eluted as a peak with a shape similar to the tritium elution curve.

- The peak in the ^{85}Kr elution curve appears to fall at a larger volume (~6 Mm^3) than the peak in the tritium elution curve (~5 Mm^3),[17] and the ^{85}Kr reaches half-maximum at 3.8 Mm^3 *vs* 3.25 Mm^3 for the tritium. Because of

the variability in the data, one cannot be sure that this is a significant difference.

- Beginning at 2 Mm^3 pumped volume (0.5 yr), the $^{85}Kr/T$ ratio at RNM-2S (see Fig. 3) has averaged 0.54 of the ratio expected for the Cambric device. Early high values (not shown) may be an artifact related to the very low concentrations of both ^{85}Kr and tritium in these samples. The 0.54 value is similar to, but possibly somewhat larger than, the ratios (0.18 to 0.51) measured in 1975 in the cavity zones at well RNM-1.

- The $^{85}Kr/T$ ratio at RNM-2S has varied during the elution; it averaged 0.41 from 2 to 6.5 Mm^3, 0.64 from 6.5 to 13 Mm^3, and 0.48 from 13 to 16.2 Mm^3 volume pumped. This variation is apparently a reflection of the delay in the ^{85}Kr elution relative to the tritium elution.

- There may be a periodic variation in the values of ^{85}Kr concentration at RNM-2S. This cycle, which appears to have a period of ~1 yr, is not readily visible in a graph of the raw data but becomes discernible when smoothing is applied. In Fig. 3, the $^{85}Kr/T$ data have been smoothed by taking an average of each point with its neighbors that are <0.2 yr away in time. The apparent cyclic variation is interesting if it is real.

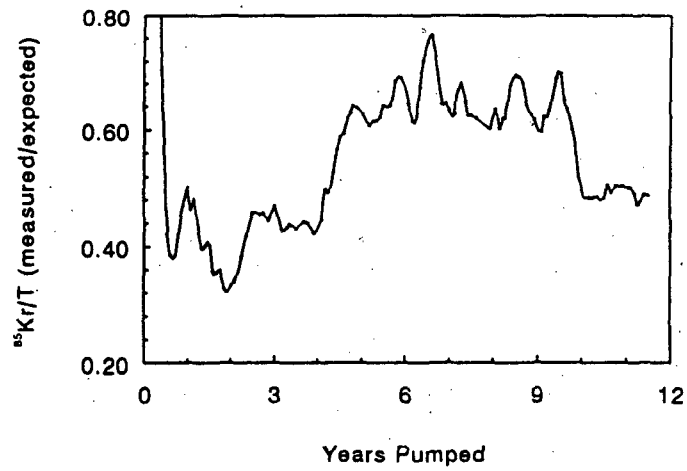

Fig. 3. $^{85}Kr/T$(measured/expected) *vs* years pumped; data have been smoothed.

In summary, ~50% of the ^{85}Kr from the Cambric explosion migrated in groundwater with the tritium, albeit somewhat more slowly. Some portion of the other 50% may be contained in fused debris, and some may have escaped to the unsaturated zone above the explosion point. Thompson[17] has suggested that the low ratios of ^{85}Kr to tritium at RNM-2S may reflect sorption or loss of ^{85}Kr in transit from the cavity to RNM-2S.

XI. ANIONIC SPECIES

Certain elements tend to form anions in aqueous solution. Anions are similar to both the noble gases and HTO with respect to their interaction with the geologic matrix. The surface charge is negative for many minerals in contact with groundwater. This negative charge repels the negative charge carried by anions,

thus keeping the anions away from the surface, and as a result, anions are not sorbed on surfaces to the same degree as cations.

Anions may also be partially excluded from certain zones that are open for the movement of groundwater. This exclusion occurs because the negative surface charge on the walls of passageways creates a repulsive field that must be overcome by an anions trying to enter the region near the wall of the passageway. Similarly, anions may be partially or totally excluded from small pores that have a negative surface charge; if this is the case, they will be concentrated in the larger pores and in the more rapidly moving water near the center of the passageways. As a result, the anions may have a greater average velocity than that of the average water molecule.

Four different anion-forming radioactive isotopes have been studied at Cambric: ^{36}Cl, ^{129}I, ^{99}Tc, and ^{106}Ru. The first is a neutron-activation product, and the last three are fission products. In all four cases, the measurements were very difficult because of the very low concentration of the species in the water.

A. Chlorine (^{36}Cl) Migration

Long-lived ^{36}Cl was formed by neutron capture in the naturally occurring chlorine (76% ^{35}Cl) present in the material around the Cambric device.[9] Although ^{36}Cl is too long lived and too low in concentration to measure by radiochemical means, its concentration can be determined by mass spectrometry. The atom ratio of $^{36}Cl/^{35}Cl$ is measured by spectrometry, and the total chloride concentration is determined by chemical analysis or by addition of a known amount of chloride tracer; these data are used to calculate the concentration of ^{36}Cl. This procedure was applied to cavity-water samples from well RNM-1 and to a series of samples pumped from well RNM-2S. The data have been reported and discussed by researchers at Los Alamos, the University of Arizona, and the University of Rochester;[9,10,15,18] a summary appears below.

The results of ^{36}Cl measurements at RNM-2S are summarized in Table III and illustrated in Fig. 4, both of which are adapted from Ref. 15. The ^{36}Cl reaches one-half of its maximum value at 2.6 Mm^3 and its maximum at ~3.8 Mm^3, putting its elution well ahead of that for tritium (half-maximum at 3.3 and peak at 4.9 Mm^3). The initial concentration in water from the cavity region was ~4.5 x 10^{11} atoms/mℓ (Refs. 9 and 11).

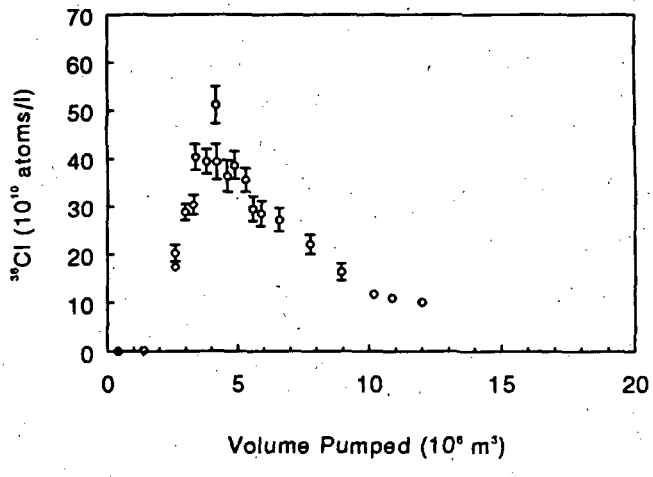

Fig. 4. Concentration of ^{36}Cl *vs* volume pumped.

Table III. Concentration of ^{36}Cl in Water from RNM-2S

Volume Pumped (10^6 m^3)	^{36}Cl Concentration (10^{11} atoms/ℓ)	Volume Pumped (10^6 m^3)	^{36}Cl Concentration (10^{11} atoms/ℓ)
0.41	0.01 ± 0.002	4.64	3.61 ± 0.33
1.40	0.03 ± 0.006	4.88	3.87 ± 0.29
2.63	1.74 ± 0.11	5.23	3.56 ± 0.25
2.61	2.03 ± 0.17	5.58	2.96 ± 0.26
2.99	2.89 ± 0.16	5.92	2.86 ± 0.26
3.18	3.04 ± 0.27	6.59	2.73 ± 0.24
3.39	4.04 ± 0.20	7.77	2.22 ± 0.20
3.81	3.95 ± 0.26	8.94	1.64 ± 0.17
4.17	5.12 ± 0.39	10.19	1.18 ± 0.10
4.20	3.94 ± 0.37	10.87	1.10 ± 0.06
4.64	3.66 ± 0.26	11.96	1.01 ± 0.05

Researchers at Los Alamos attributed the early elution of ^{36}Cl to the anion exclusion effect.[15,18] (See the section on hydrological modeling for further discussion of the ^{36}Cl data.)

B. Iodine (^{129}I) Migration

The anion forming radionuclide ^{129}I also was measured in water samples from RNM-2S.[3,9,10,11,12] Because of its long half-life (2 x 10^5 yr) and low concentration, the fission product ^{129}I cannot be measured by radiochemical means. Instead, it was measured by neutron activation and tandem accelerator based mass spectrometry (TAMS).

The results of a series of analyses are shown in Fig. 5 and Table IV . As was the case with ^{85}Kr, there is a large amount of scatter in the data. It is not clear whether this is real or an artifact of the analyses.

Because of its many oxidation states and its chemical reactivity, the behavior of iodine at very low concentrations is complex[19] and difficult to predict, and iodine analyses are sometimes plagued by "analytical problems." It should not come as a surprise if iodine's migration behavior were complex. At the pH-Eh conditions prevalent at Cambric, some of the iodine could exist as HIO, a weak acid. It has been suggested that this reactive species may interact with matrix materials.[10]

The ^{129}I elution may begin before the tritium elution; the rising portion of the ^{129}I curve extrapolates to zero concentration at a volume of 1.5 Mm^3 *vs* 2.0 Mm^3

for tritium. It is not entirely clear from the data whether a peak in concentration had been reached by the time of the last sample. The atom ratio of ^{129}I to tritium was ~0.0003 near the tritium peak. Two measurements of ^{129}I concentration at RNM-1 indicated that the ^{129}I-to-tritium ratio was ~0.005. The expected ratio for the Cambric source is 0.0012.

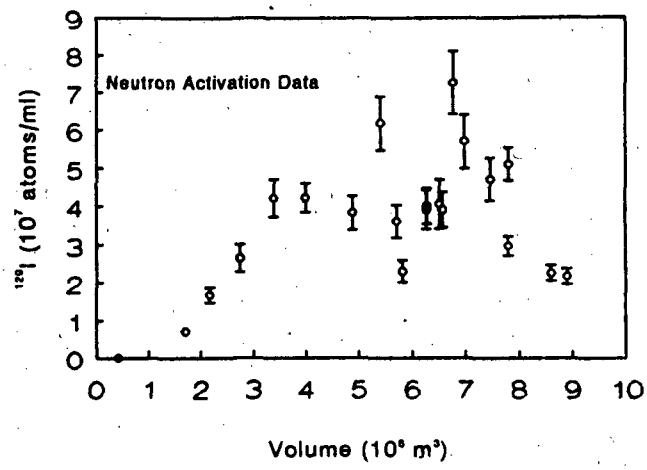

Fig. 5. Concentration of ^{129}I *vs* volume pumped.

A substantial fraction of the ^{129}I migrated from the Cambric site to RNM-2S. However, some of the iodine is immobile or its migration is retarded to some extent (perhaps it is all mobile and the variability in results is an indication of basic problems in the sampling and analysis).

C. Technetium (^{99}Tc) Migration

The long-lived (2×10^5 yr) fission product ^{99}Tc, like ^{129}I, is difficult to measure by radiochemical means but can be measured by mass spectrometry. It also can exist in several oxidation states; in the oxygenated water at the Cambric site, it is expected to form the pertechnetate anion. As an anion, it should be free to migrate.

Ogard and Rokop[17] and Schroeder and Rokop* assayed water samples from both RNM-1 and RNM-2S for ^{99}Tc by a mass spectrometric method; the results are shown in Table V. The measured ratio of ^{99}Tc to tritium indicated that only ~0.01% of the ^{99}Tc was in solution. The ratio to tritium is rather similar for the two wells, suggesting that the soluble ^{99}Tc migrated freely with the tritium. By analogy with other nuclear tests, it is certain that a substantial fraction of the ^{99}Tc is immobilized in glassy debris. There is insufficient data to draw any conclusions about the elution peak arrival time or shape.

D. Ruthenium (^{106}Ru) Migration

With a half-life of only 1 yr, the fission product ^{106}Ru had mostly decayed to stable ^{106}Rh by the start of the Cambric migration experiment. Nevertheless, there was a sufficient quantity left to permit measurement of samples from RNM-1 and RNM-2S by radiochemical means.

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* The ^{99}Tc data for RNM-2S were provided by N. Schroeder and D. Rokop, Los Alamos National Laboratory, Los Alamos, NM 87545.

Table IV. Concentration of ^{129}I in Water Samples from RNM-1 and RNM-2S

Sample	Volume Pumped at RNM-2S[a] (Mm^3)	^{129}I Concentration[b] (atoms/mℓ x 10^{-7})	$^{129}I/T$ (atom ratio)
Well RNM-1			
Cavity			
Lower	0, (2)	14100.	1.21×10^{-3}
Lower	0, (22)	21400.	1.23×10^{-3}
Upper	0, (14&18)	7700.	5.6×10^{-4}
Chimney	0, (27&41)	12.1	4.2×10^{-5}
Periphery	0, (19)	4.2	4.2×10^{-5}
Chimney	1.17	0.54	8.7×10^{-5}
Chimney	1.34	0.59	1.4×10^{-4}
Well RNM-2S			
	0.4	0.57	6.7
	1.4 - 1.9	1.0	0.019
	2.2 - 2.6	2.0	0.00075
	3.3 - 3.9	4.5	0.00049
	4.2 - 4.9	4.2	0.00030
	5.2 - 5.8	5.4	0.00038
	6.3 - 7.0	4.8	0.00037
	7.5	5.0	0.00043

[a] Values in parentheses represent total cubic meters of water pumped from the particular zone of RNM-1 before sample collection.
[b] Values for RNM-2S samples were averaged over the indicated range of volume pumped.

Ruthenium exhibits complex chemical behavior as a result of its strong ability to form complex ions and its multiple oxidation states. Isotopic exchange among the different complex ions is often slow. Some of the complex ions carry a negative charge and thus are expected to migrate readily.

Analyses of Cambric samples for ^{106}Ru were conducted at Lawrence Livermore National Laboratory (LLNL) and also at Los Alamos. Coles and Ramspott[20] reported results from analyses of a series of 200-ℓ water samples collected from RNM-2S from 1978 to 1980. The ^{106}Ru concentration ranged from 0.04 to 0.08 pCi/ℓ

Table V. Concentration of ^{99}Tc in Water Samples from RNM-1 and RNM-2S

Sample Location	Volume Pumped - RNM-2S (Mm^3)	^{99}Tc Concentration (atoms/mℓ)
RNM-1, cavity area	0.	8.15×10^7
RNM-2S	4.9	8.46×10^4
	5.2	1.24×10^5
	6.0	1.02×10^5

when measured 15 yr after the explosion; the higher concentrations first appeared in samples collected in August 1979. These very low concentrations were difficult to measure but were confirmed by running a blank (<0.02 pCi/ℓ) and by parallel analyses carried out at Los Alamos.

The authors pointed out that the initial ^{106}Ru concentration of ~120 pCi/ℓ in water from well RNM-1 had disappeared during 4 yr of pumping and suggested that this indicated that the ^{106}Ru was mobile, had been swept from the cavity region, and had not been replenished by leaching from melt glass. It was noted that in laboratory experiments, ruthenium has always exhibited a significant tendency to sorb on rock (K_d = 10 to 8000) and that the 91-m migration observed at Cambric would require a $K_d \leq 0.3$ mℓ/g. They concluded that the migration probably reflects ruthenium's complex chemical behavior and that the migrating species was probably RuO_4^{2-}. The authors made the important point that sorption behavior seen in laboratory experiments is not always directly applicable to field conditions.

From the $^{106}Ru/T$ ratios it appears that only ~1% of the Cambric ^{106}Ru was in the cavity water.[2] The rest was either adsorbed or contained in the melt glass. The $^{106}Ru/T$ ratios at RNM-2S were similar to the initial values at RNM-1 near the cavity, which suggests that they migrated at similar rates.

XII. MIGRATION OF OTHER RADIONUCLIDES

Radionuclides that form cationic species in aqueous solution are likely to be sorbed on the minerals in the alluvium surrounding the Cambric test. Analyses for two cationic radionuclides, ^{90}Sr and ^{137}Cs, were carried out on samples collected from RNM-1 and RNM-2S at various times during the Cambric migration experiment. RNM-1 was generally sampled once each year, whereas samples were collected more frequently (normally once each month) from RNM-2S.

Gamma-ray spectroscopy was used to measure radiations from the residual solids produced by evaporation of 200-ℓ samples, and radiochemical separation and purification procedures for ^{90}Sr and ^{137}Cs were applied to 2- or 4-ℓ water samples from RNM-2S. The fission product ^{137}Cs, with its 662-keV gamma ray, could be detected if present in the 200-ℓ samples at a concentration of ~1×10^3 atoms/mℓ (0.02 pCi/ℓ). The fission product ^{90}Sr, with no gamma rays, is more difficult to assay and would be detectable in the 2-ℓ samples at a concentration of ~5×10^5 atoms/mℓ (10 pCi/ℓ) (Ref. 13).

Table VI. Concentration of ^{90}Sr and ^{137}Cs in Water from the Chimney Area of Well RNM-1

Volume Pumped RNM-2S (Mm^3)	^{90}Sr (pCi/mℓ)	^{137}Cs (pCi/mℓ)	$^{90}Sr/T^a$ (x 10^4)	$^{137}Cs/T^a$ (x 10^4)
0.	2.7	0.045	0.4	0.07
0.[b]	0.11	0.090	0.07	0.06
1.17	0.46	0.11	3	0.8
1.34	0.27	0.22	3	3
3.50	0.091	0.045	8	4
5.89	0.031	0.026	5	5
6.48	0.027	0.030	8	9
7.67	na	0.026	na	19
8.91	0.017	0.020	18	22
10.0	0.011	0.020	20	38
11.1	0.018	0.020	72	100
12.2	0.003	0.019	20	130

[a] Atom ratio at zero time.
[b] Periphery.

Results from analyses of RNM-1 samples are summarized in Table VI, which was adapted from Ref. 15. Reports for analyses of RNM-2S samples have been limited to the statement that the isotopes have not been detected; this implies that the concentration of ^{90}Sr was below ~10 pCi/mℓ and that the concentration of ^{137}Cs was below ~0.02 pCi/mℓ at RNM-2S when the samples were collected. No details were reported.

In addition to the ^{90}Sr and ^{137}Cs analyses, one ^{239}Pu analysis was carried out on a water sample collected on May 7, 1986 from RNM-1 and one on a 200-ℓ sample collected on June 17, 1981 from RNM-2S shortly after the tritium peak.* The concentration in both instances was below the normal 10^6 atoms/mℓ detection limit.** The sample from RNM-2S was assayed for ^{241}Am as well and found to contain less than 10^5 atoms/mℓ.

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* Information received from J. L. Thompson, Los Alamos National Laboratory, Los Alamos, NM 87545 (July 26, 1991).

** For the RNM-1 sample, the blank-corrected results reported were negative (-2 x 10^5 atoms/mℓ); for the RNM-2S sample, the results were reported as <4.25 x 10^4 atoms/mℓ for the sample and <4.73 x 10^4 atoms/mℓ for the accompanying blank.

XIII. PARTICULATE STUDIES

Transport of materials by groundwater is generally assumed to refer to materials dissolved in the groundwater. Based on many observations, it appears that under certain conditions particulates can be transported as well. Stable suspensions of particles, called colloidal suspensions, are fairly common in nature; two examples are humic substances and clay particles. Both of these materials are chemically active and are able to strongly bind radionuclides such as ^{239}Pu and ^{241}Am. The colloidal particles with the radionuclides bound to them can then be transported by groundwater.

There are several often-cited examples of colloidal transport; one is the LLNL interpretation of data from the Cheshire site at the NTS.[21] At Los Alamos, researchers reported that americium had migrated a considerable distance from a waste-stream out-fall – much farther than anticipated; they presented evidence that the americium is in particulate form instead of being in solution.[22] Researchers at Chalk River Nuclear Laboratories in Canada have reported that cesium migrated away from buried test blocks of nuclear waste more rapidly than anticipated and concluded that it had moved in particulate form.[23]

There has been some interest in determining the nature of colloidal suspensions that may be present in groundwater at Cambric. In particular, one would like to know whether any of the particles present in the groundwater are capable of adsorbing any of the radionuclides at Cambric and whether any of the particles at Cambric have migrated toward or to the RNM-2S well.

In 1983, a 300-ℓ volume of water from RNM-1 was filtered through 0.4-μm-pore filters and a 3770-ℓ volume from RNM-2S was filtered through 0.05-μm-pore filters. Particulates studied with a scanning electron microscope (SEM) contained a variety of different elemental compositions – predominantly silicon. It was reported that particle diameters ranged from <0.001 μm to tens of micrometers.[11]

S. Goldstein and J. Fabryka-Martin reported on the character of particulates at RNM-1 (Ref. 17). In 1988, an 80-ℓ water sample from the chimney zone of well RNM-1 was brought to Los Alamos in a polyethylene bottle and was subsequently processed with a Millipore Pellicon ultrafiltration system (100 000-NMWL filter) to produce a 250-mℓ concentrate. Portions of the concentrate, which was noticeably cloudy, were centrifuged onto carbon films that were supported on copper grids for transmission electron microscopy (TEM) analysis. A blank was processed in a similar manner and proved to be quite clean.

The TEM samples were too concentrated for good resolution of individual particles, but there were both amorphous and crystalline particles present. Results of the TEM assay appear in Table VII. The crystalline particles were tentatively identified as clay, whereas the amorphous material seemed to be similar to apatite (a phosphate). Both the clay and the amorphous material might have been introduced as drilling materials. The natural phosphate concentration is quite low in NTS waters.

In December 1990, only a week after the pump at RNM-2S had been restarted, E. Bryant and D. Rokop used filter apparatus at RNM-2S to collect size fractions for weighing and to concentrate particulates from a fairly large volume of water

while ensuring minimal chance for chemical changes during the process. The concentrate was to be used for characterization studies.

Several filter stacks with pore sizes from 8- to 0.015-μm diameter were used. Unfortunately, the data were compromised by experimental difficulties. Reduction in flow rate for the 1- and 0.1-μm filters indicated the presence of particulates in the water; however, the maximum weight gain for any of the filters was 0.6 mg, and some of that was apparently caused by growth of algae on the filter substrate during two long, unplanned storage periods at NTS. It was not possible to achieve a sustained flow through the smallest filter, and this was later attributed to clogging by fine particles that formed when the water was cooled by the winter air. The large-size filter stacks were subsequently dried and weighed along with a blank stack. The measured particle concentrations were very low for all particle sizes, but a gain in weight was observed for all the filter membranes, including a stack of blanks. The blank-corrected loadings (0.01, -0.005, and 0.01 mg/ℓ for 0.1- to 1-μm, 1- to 8-μm, and >8-μm particles, respectively) were smaller than the 0.05-mg/ℓ standard deviation for all weight gains. These very low loading values are interesting because it has often been suggested that rapid pumping might generate particles; in this case, there seem to have been very few generated particles.

SEM was used to examine particulates from the RNM-2S 100-to-1 concentrate of particles greater than 300 000 NMWL. Attempts to examine submicrometer particles directly on the filter substrates were unsuccessful as a result of poor resolution. SEM pictures for a sample evaporated on a carbon substrate were of good quality and showed that particles in the 0.1- to 10-μm range were mostly calcium minerals (probably calcite, the carbon and oxygen are invisible to SEM) and occasional silica particles; other agglomerates contained a mixture of cations (calcium, sodium, potassium, and silicon) and anions (sulfur and chlorine). In addition, there were highly crystalline calcite particles that appeared to have recently precipitated; these particles differed sharply in appearance from the generally rounded calcium mineral particles mentioned in the previous sentence.

Photon correlation spectroscopy was applied to a series of samples from the above concentrate. In some cases, the aliquot was first filtered through a Nuclepore filter to remove larger particles. Results indicated (1) the presence of particles with a wide range of sizes but concentrated in the 0.1- to 1-μm range, and (2) the transient presence of very small (~1-nm) particles.

The clogging of filters in the cold December air at the NTS and the appearance of transient fine particles at Los Alamos are probably both a reflection of the dynamic equilibrium between particles and solution. At Los Alamos, the particle concentrate had been stored in a clean-room that was exceptionally warm. Apparently, the warm water dissolved some of the particulate material, which precipitated again when it was removed from the warm area. The fine particles had disappeared from the suspension (presumably by recrystallization to larger particles) by the second day after the samples were removed from the clean area.

The geochemistry speciation code PHREEQE was used to calculate saturation indices for water from well 5B, which is not far from the Cambric site. As indicated in Table VIII, a number of minerals had positive indices, indicating the potential for particle precipitation.[17]

Table VII. RNM-1 Particle Analysis by TEM

Sample	Major Elements	Minor Elements	Structure
General area scan	Si	Ca,S,P,K,Fe	
Particle	Ca, P	Mg	Amorphous, 0.05-0.1 μm
Nearby material	Si,Ca,P,S	Fe,Mg	Amorphous
Particle	Mn,Si	Ca,S	Amorphous, 0.05-0.1 μm
Particle	Ca,P	Mg	Amorphous, 0.3-0.4 μm.
Large particle	Si,K,Al	Na	Crystalline, 0.5 μm, blocky
Nearby material	Si,Al,Mg	Fe,K	Amorphous
Particle	Si,Al	Ca,Fe,K	Crystalline, 0.5 μm
Particle	Si,Al,K	Ca,Fe	Crystalline
Particle	Ca,P	Mg	Amorphous, 0.3 μm
Particle	Ca,P		Amorphous, 0.15 μm
Particle	Ca,As	Si,Mg?	Amorphous, <0.1 μm

XIV. HYDROLOGICAL MODELING

Three groups have carried out model calculations for the Cambric migration experiment. In each case, the primary focus has been to simulate the elution of tritium or ^{36}Cl in the water pumped from RNM-2S, and in each case, the limiting factor has been information about the hydraulic parameters in the area of the Cambric site. In this section, we first look at the problem in a very simple way and then pass on to more sophisticated analyses.

A. Back-of-the-Envelope Model

The simplest model would assume a confined aquifer in a uniform, isotropic medium. In such a system, one might estimate the travel time for water from RNM-1 to RNM-2S by calculating the volume of water contained in a cylinder whose axis is at RNM-2S and perimeter is at RNM-1, 91 m away. If one ignores dispersion, the travel time would be the water volume divided by the pumping rate. Using a porosity of 0.3, a radius of 91 m, and a cylinder height of 206 m (twice the distance from the water table to the perforations at RNM-2S), one obtains a volume of 1.61 Mm^3. At this volume, one might expect to have pumped out ~50% of the tritium. In fact, the tritium concentration at RNM-2S had reached only ~0.1% of its peak concentration at this volume and the peak arrived at 4.9 Mm^3. Clearly, the hydrology at the Cambric site is not this simple.

Table VIII. Saturation Indices for Water from Well 5B, Frenchman Flat, NTS

Mineral	Saturation Index	Mineral Formula
Apatites		
Carbonate apatite	12.8	$Ca_{9.5}\ Mg_{0.14}\ F_{2.48}\ (PO_4)_{4.8}\ (CO_3)_{1.2}$
Hydroxylapatite	-1.5	$Ca_5\ (PO_4)_3\ OH$
Fluorapatite	7.6	$Ca_5\ F\ (PO_4)_3$
Feldspars		
Microcline (K-spar)	4.4	$KAlSi_3O_8$
Sanidine	3.9	$KAlSi_3O_8$
Anorthite	-0.2	$CaAl_2Si_2O_8$
Micas		
Muscovite (K-mica)	10.2	$KAl_2(AlSi_3O_{10})(OH)_2$
Pyrophyllite	7.9	$Al_2Si_4O_{10}(OH)_2$
Zeolite		
Laumontite	4.8	$(Ca,Na)_7Al_{12}(Al,Si)_2Si_{26}O_{80}\cdot 25H_2O$
Clays		
Kaolinite	6.5	$Al_4(Si_4O_{10})(OH)_8$
Nontronites	21-28	$Fe_4AlSi_8O_{20}(OH)_4$
Illite	4.4	Mica-like clay mineral
Montmorillonite	3.9	$(Al,Mg)_8(Si_4O_{10})_3(OH)_{10}\cdot 12H_2O$
Chlorites	5-31	$(Mg,Fe)_3Si_4O_{10}(OH)_2Mg_3(OH)_6$

B. Sauty Model

A more complex model is based on Sauty's discussion of hydrodispersive transfer in aquifers.[24] Sauty provided a series of calculated "type curves" with dimensionless variables for several different geometries. The curves for radially convergent flow are applicable to confined aquifers with uniform properties. To use these curves, one converts the measured data to dimensionless form. Concentration is divided by maximum concentration to obtain a dimensionless concentration ratio. To obtain a dimensionless time, one divides the volume pumped by the pore volume of a cylinder whose axis is at the pumped well and outer radius is at the tracer injection point and whose height is equal to the vertical thickness of the confined aquifer. If the cylinder volume is unknown, one prepares a plot with the pumping time on a logarithmic axis and the concentration on a linear axis. The only remaining parameter is the Peclet number, which is the ratio of the radius of the cylinder to the longitudinal

dispersivity. The Peclet number is chosen to obtain the best agreement between the shape of the "type curves" and the plot of experimental data. Sauty recommends using the semilog plot and sliding a similar plot of the real data along the time axis until a match is found for the curve shape. The information obtained from such a fit to the Sauty curves is a value for dispersivity.

The Sauty model has been applied to the Cambric data several times. For Cambric, the cylinder radius is 91 m, but the height is not well defined and the initial dispersion of tritium does not match the point injection assumed for the calculation. There are questions about the validity of this one-dimensional model for the 3-dimensional Cambric geometry, as was pointed out by Burbey and Wheatcraft.[7] They estimated 2100 days for the time required to pump out the cylinder and derived a Peclet number of 45 that was based on fitting the time to breakthrough and to the peak of the tritium concentration; the corresponding dispersivity is 2 m (= 91 / 45).

Travis et al.[25] reported a good fit for data up to 6 Mm^3 pumped volume using a Peclet number of 10, but the tritium source had to be adjusted downward to 70% of the original estimated value.

Ogard et al.[15,18] reported that for tritium elution data up to 12 Mm^3 volume pumped, a Peclet number of 6 gave a better fit than did a value of 10.

Application of the Sauty model to Cambric has produced estimates for dispersivity that range from 2 to 15 m. This model is probably not appropriate for the Cambric experiment, but it is simple and permits a good fit to the shape of the elution.

C. Selection of an Appropriate Model

The focus of a report by Wheeler et al.[6] was the selection of suitable hydrologic models for treating the Cambric data. They concluded that the model selection criteria indicated the need for "a fully 3-dimensional computer model." They further stated that the model should simulate confined and unconfined aquifers and should handle sorption-desorption of radionuclides. Four existing, previously reviewed[26] computer codes were recommended: DAVIS/FE3D, MAIN, PINDER, and SWIFT. During the selection process, the authors carried out a number of calculations with relevant results. The following paragraphs review some of the information and results from this report.

The Cambric site lies in a layer of tuffaceous alluvium at least 500 m thick. Below the water table, there is evidence of weathering to fine argillaceous material with low permeability. The alluvium is probably locally layered by the stream-bed deposition mechanism, but no single layer can be expected to extend over long distances. Fine-grain deposits may serve as aquitards, or confining layers, to restrict the vertical flow of water on a local scale. Vertical conductivity is probably ~0.1 of the horizontal, on average, but with local variations.

The water inlet perforations at RNM-2S are well below Zone-IV at RNM-1 (325- to 350-m *vs* 273 to 281-m depth) and thus require a vertical flow component to transport tritium from RNM-1 to RNM-2S. A fine, silty clay layer, possibly

capable of slowing the flow of water, was identified in this interval at 292-m depth in well U5e; perhaps fortuitously, a similar layer was observed at the same depth in RNM-1. The existence of such a low-permeability layer could have important consequences for the movement of water from the Cambric test point to the perforations at RNM-2S.

Aquifer properties were derived by analysis of data collected at RNM-2S during pumping from RNM-1 as well as data collected at RNM-1, RNM-2, and RNM-2S while pumping at RNM-2S. Order of magnitude, model-dependent values were obtained for the storage coefficient ($\sim 10^{-3}$), transmissivity ($\sim 10^2$ m^2/d), effective aquifer thickness (~20 m), and permeability (~3 m/d). (The lowest permeability values observed were those reported by Hoffman, Stone, and Dudley,[2] which were ~0.2 m/d for the cavity area (Zones II & III).)

There appeared to be little or no effect from pumping RNM-2S on the drawdown in the chimney area at RNM-1 (Zone IV). In contrast, an 8-hr pump test at RNM-1 during July 1974 apparently produced a drawdown of 0.3 m at RNM-2S; however, as discussed below, there is some doubt about the validity of this observation. Based on the apparent lack of drawdown, one would conclude that the communication between RNM-1 and RNM-2S is somewhat limited.

Pressure oscillations with an amplitude of ~1 m drawdown were seen in the chimney area (Zone IV) of well RNM-1. These appeared to be diurnal in nature and seemed to reflect the effect of local temperature variations on barometric pressure. Similar oscillations in pressure were seen at RNM-2S. Such oscillations can create problems in the interpretation of drawdown data, especially if the timespan of the drawdown measurement is a significant fraction of the 24-hr cycle time. These observations cast some doubt on the validity of the data from the 8-hr drawdown test at RNM-1 and, consequently, on the derived parameters.

Based on their analysis of the data, Wheeler et al. concluded that (1) the aquifer responds as a confined, leaky-artesian aquifer with a vertical-to-horizontal permeability ratio of 1 to 10, and (2) the hydrologic system is too complex to be evaluated by analytical methods, but a simulation model can be sufficiently accurate to describe the results of the Cambric migration experiment.

The US Geological Survey Method of Characteristics Model for Solute Transport in a Porous Medium[27] (SOLTRN) was used to simulate the two pumping tests at the Cambric site. Parameters, selected to obtain a good fit, are shown in Table IX.

The model used the derived properties, including the 10:1 ratio of horizontal to vertical conductivity, and assumed that tritiated water is an ideal tracer to predict a peak tritium concentration of 3.8×10^4 pCi/mℓ at 0.8-Mm^3 volume pumped. The observed peak was 7×10^3 pCi/mℓ at 4.8 Mm^3.

The model was then used to indicate sensitivity to parameter values. A factor of 100 reduction in the horizontal permeability in the cavity zone and factor of 2 increase in the dispersion coefficient lowered the predicted peak concentration to 1.5×10^4 pCi/mℓ at 1.0 Mm^3. A general factor of 4 reduction in horizontal permeability and a factor of 5 increase in vertical permeability resulted in a predicted peak concentration of 1.3×10^4 pCi/mℓ at 2 Mm^3.

Table IX. Aquifer Properties Selected by SOLTRN Simulation

Depth Below Water Table (m)	Permeability (m/d)
0 - 100	3.4
100 - 125	1.4
125 - 200	0.7
Ratio of vertical to horizontal permeability	0.1
Storage coefficient	0.0001
Leakage from water table	90% of pump rate
Leakage from below	1% of pump rate
Leakage from lateral inflow	9% of pump rate

None of these predictions could be considered to be a satisfactory fit to the data. Failure to predict the observed tritium elution was attributed to the three-dimensional heterogeneity of the aquifer properties.

It may be noted that the observed peak is about six times lower and appears at a volume about six times greater than is predicted by the simulation when derived parameters are used. From this, one might infer that the flow rate from the cavity to RNM-2S was roughly six times smaller than the average flow rate for all the water being pumped at RNM-2S.

D. TRACR3D Model

Travis et al.[10] employed the code TRACR3D[28] to simulate the elution of tritium from RNM-2S. The simulation assumed four homogeneous hydrologic environments: the general aquifer, the "cavity," the surrounding hemispherical crushed zone, and the gravel pack around the pipe at well RNM-2S. The properties assumed for these zones are listed in Table X. Initial tritium concentrations were 3.8 and 6.1 μCi/mℓ in the upper and lower "cavity," and a background of 0.25 pCi/mℓ was assumed.

To reduce the time required for the calculation, only a 14° pie-slice of the Cambric geometry was included. *This slice is just wide enough to include the 22-m-diameter Cambric "cavity" but not all of the 5-m "crush zone" assumed to surround the cavity zone. It is not clear whether this exclusion had a significant effect on the results. The wedge was 200 m in height and its top was at the water table.*

The simulation predicted breakthrough at 1.5 and peak concentration at ~6 Mm^3 – somewhat early but in rough agreement with observation.

Table X. Parameters Used for the TRACR3D Simulation

	Alluvium	Crush-Up	Cavity	Gravel
Permeability (Darcys)	1.0	0.05	1.0	50.
Porosity				
intergrain	0.38	0.10	0.45	0.40
intragrain	0.06	0.06	0.06	0.0
Tortuosity factor	0.3	0.15	0.30	1.0

E. Deep Well Disposal Model

In their report on the Cambric experiment, Burbey and Wheatcraft[7] presented results from an extensive modeling effort. Their first task was to assign values to parameters for the calculations. They then carried out simulations of both the tritium and the ^{36}Cl data by using the fully transient, 3-dimensional, finite difference "Deep-Well Disposal" model.[29] This model includes provision for sorptive interaction of tracers with the matrix.

Most hydraulic and dispersive parameters were based on analysis of field data and were held constant in the model. Variable parameters were the hydraulic conductivity and porosity of the cavity, the vertical-to-horizontal hydraulic-conductivity ratio, transverse dispersivity, and the retardation coefficient. These parameters were varied in a series of simulations to show sensitivity of the results to the parameter values and to obtain a best fit to the data.

Particular attention was given to the initial distribution of tritium. Using statistical analysis of the Cambric data, they chose 6.93 μCi/mℓ as the best value for tritium concentration within a tritiated radius of 18 m. As indicated previously, they used the Sauty model to choose a dispersivity of 2.0 m.

In applying the model to the Cambric geometry, Burbey and Wheatcraft employed a square block of cells in which Cambric was near one corner and RNM-2S was at the opposite corner. The top of the block was 39 m below the water table and the block extended downward for 105 m. Conductivities were assumed to be uniform horizontally but varied in the vertical direction. Using analysis of data from hole Ue5n, they assigned relatively low values for conductivity (0.20 and 0.28 m/d) to two strata at and just below the cavity.[30] Values of 2.5 to 3.0 m/d were assigned to layers further below the cavity, and a value of 0.25 was assigned to the "cavity" cell itself.

The selected values (K_h/K_v = 5.0, α_T = 1.5 m, K_{cav} = 0.25 m/d, and ϕ_{cav} = 0.15) provided a reasonably good fit to the shape of the tritium elution, but it was ~200 to 300 days early. The only model parameter left to delay the elution was the absorption parameter K_d. A value of 0.8 mℓ/g was used to provide the needed

delay in the simulated tritium elution. A simulated elution of ^{36}Cl is discussed below in the section on anion exclusion.

Two comments should be made regarding the use of tritium sorption in the simulation. First, even with no sorption the fit was quite good; it peaked ~300 days early out of 2100 days and was only slightly high in concentration. The inclusion of sorption can be viewed as fine-tuning; a sorption value based on fine-tuning of the simulation of a poorly defined system is itself not well defined. Second, the magnitude of the reported K_d (0.8 mℓ/g) is puzzling because such a value would imply that only ~20% of the tritium would be mobile at any given instant. The result should be a five-fold decrease in average tritium velocity instead of a 15% reduction.

Three possible mechanisms for the "delay" of tritium were suggested: (1) adsorption of tritiated water molecules into the film surrounding the clay particles, (2) a "self-diffusion" process wherein tritium is diffusing into immobile water regions, and (3) exchange with the structural water of the clay. "Self diffusion effects" are described as the diffusion of tritiated water into stagnant water pockets (such as pores or films) or the exchange of tritiated water molecules with chemically bound water in the matrix.

It may be noted in passing that for a given total water content, the simple exchange of mobile water with immobile water (with no isotope effect) produces dispersion but does not lower the average velocity for the water or tritiated water as a whole, relative to the average velocity for 100% mobile water. To delay the elution by exchange or self-diffusion, one must introduce additional immobile water.

Based on the results of their modeling, Burbey and Wheatcraft came to the conclusions listed below regarding the potential for migration away from nuclear detonation sites.

- The hydraulic conductivity of the cavity was low, resulting in a reduction in the peak concentration at the sampling well.
- The simulated migration of tritium was relatively insensitive to cavity porosity, vertical conductivity, and transverse dispersivity.
- It was necessary to use retardation to match the tritium elution curve; the value used agrees with values found in the literature.
- It was necessary to use retardation to match the ^{36}Cl elution curve; it is not clear whether ^{36}Cl is being advanced or retarded relative to the advection front.
- Late-time deviation of the simulated tritium elution curve from the measured curve may reflect the recirculation of tritium from the ditch where the water pumped from RNM-2S is dumped, or it may indicate a complex initial tritium source distribution.

F. Anion Exclusion

As was mentioned previously, the ^{36}Cl elution at RNM-2S preceded the tritium elution by a significant volume and researchers at Los Alamos attributed this phenomenon to the "anion exclusion" effect. From parameters selected to fit both the ^{36}Cl and the tritium elution curves with the ideal curves provided by Sauty,[24] they were able to estimate an "exclusion volume" of 7 mℓ/100g of alluvium, which is comparable to values reported by Thomas and Swoboda.[31] A reasonable fit to the shape of ^{36}Cl elution was obtained with a Peclet number of 6, which corresponds to a dispersivity of 15 m for the Cambric geometry.

Burbey and Wheatcraft[7] simulated the ^{36}Cl elution curve with the same set of hydraulic parameters selected for use with tritium. A smaller radius was used for the initial ^{36}Cl distribution than for the initial tritium distribution (8.5-m *vs* 18-m radius), and a K_d value of 0.5 mℓ/g was used for ^{36}Cl *vs* 0.8 mℓ/g for tritium.*

Using the rather good fit of the simulation to the data, Burbey and Wheatcraft concluded that the anion exclusion effect may be dominated by "self diffusion effects" that act to retard the advancing front. ***However, the reduction in K_d to 0.5 mℓ/g might be considered to constitute a** de facto **"exclusion effect."***

XV. INFILTRATION AND RECHARGE STUDY

The water pumped from RNM-2S is dumped into a ditch a short distance from the well; it flows along the ditch for ~1 mile and then into a pond. Researchers at LLNL and the Desert Research Institute (DRI) have studied the infiltration and recharge of the tritiated water into the vadose zone along and below the ditch.[32,33] The approach and findings for the recharge study are summarized here.

A series of three flumes was installed along the ditch. DRI personnel used the measured flow rates and estimates for maximum evapotranspiration rates to estimate that infiltration losses through the bottom of the ditch amounted to 175 gpm between the first and third flumes. This corresponds to a piston flow pore velocity of ~73 m/yr at 25% porosity. R. Jacobson, from DRI, suggested that the recharge plume had reached the water table after 9 yr of pumping.

Soil samples were collected from a series of 8- to 15-ft-deep holes near the ditch, and instruments (suction lysimeters, tensiometers, and resistance cells) were installed in the same holes. The soil moisture increased with depth next to the berm and also at a point 11 ft from the berm; at 21 ft from the berm, only ambient (dry) moisture levels were encountered. Similarly, tritiated water, at or above the ditch concentration, was encountered in the wet and moist zones next to and at 5 feet from the berm. At 11 ft from the berm, tritium concentration increased with depth from 5% at 5 to 10 ft to 50% of the ditch value at 15-ft depth. At 21 ft from the berm, the tritium concentration was ≤0.1% of the ditch level down to 10-ft depth.

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* See the section on hydrology for comments on the use of a K_d for tritium.

The tritium content of the ditch water and water from the lysimeters was followed from late 1983 to the middle of 1985. During this period, the tritium concentration in the ditch dropped steadily. The tritium concentration measured in lysimeter water from a depth of 12 ft, 5 ft from the berm, paralleled the drop in the concentration in the ditch but was ~25% larger in magnitude at any given time. This finding was attributed to the delay time required for water to flow from the ditch to the lysimeter position. A lag time of 1.5 yr provided excellent agreement between calculated and observed tritium values. Similarly, lysimeter water samples collected even further from the ditch were subject to an even longer lag time and reflected the rising tritium levels in the ditch some 6 yr before the sampling dates.

A 30.5-m borehole was drilled and sampled in May 1985, 2.4 m from the berm and downstream from the previous array of sample holes. Core samples were all composed of coarse, highly permeable material and were unsaturated. Tritium concentrations were uniform and equal to recent values in the ditch, which was consistent with a transport time of no more than 4 months. A bromide tracer experiment was performed to determine the actual transport time from a short section of ditch to a lysimeter at the bottom of this hole. The peak in the bromide concentration at the lysimeter appeared at 92 days after injection, which corresponded to a transport velocity of ~0.3 m/d.

The LLNL researchers pointed out the potential value of the Cambric ditch and underlying zones as an opportunity to study the movement of water and radionuclides in alluvium as a result of gravity-driven flow and lateral, unsaturated migration in the vadose zone.[33] Current research at the Cambric ditch makes use of ^{3}He as an indicator of the migration time for water from the ditch to the sampling point, based on the half-life for the decay of tritium to ^{3}He.[34]

XVI. VEGETATION WATER BUDGETS

Ruggieri, Buddemeier, and Jacobson[35] reported the results from a study of tritium and deuterium in plant materials collected along the Cambric ditch and the pond into which it empties. An extensive set of data appears in their report. They observed that for water in plant materials from or near the ditch, deuterium was generally slightly enriched, whereas tritium in these materials was as much as 70% depleted relative to ditch water. They concluded that the depletion was caused by the exchange of water between plant and atmosphere. The atmosphere was the source of 50% or more of the water in the plants at night, but during the day, the tritium content of the plant water rose until it corresponded to ~90% that of ditch water. The small effects for deuterium were a reflection of the small differences in deuterium enrichment among the water sources. Although it was surprising that the atmosphere would be the source of such a large fraction of the plant water at low ambient humidity values, these observations were in accord with results of similar laboratory experiments and indicated rapid plant-atmosphere exchange rates.

XVII. SUMMARY

A. Hydrologic Modeling

A variety of models have been applied, each with its own particular merits and limitations. The degree to which each model was able to provide a fit to the tritium elution reflected both its suitability and the parameters selected for use with the model. A common theme was that the actual elution was delayed relative to the elution predicted for an isotropic medium. This delay was variously accommodated by (1) introducing a reduced permeability for the cavity and/or the surrounding volume, (2) including a horizontal layer with low conductivity between the cavity and the pump perforations, and (3) allowing tritiated water to be "adsorbed" by the matrix.

The paucity of detailed information about the physical nature of the pathway from the source at RNM-1 to the sink at RNM-2 is a normal and unavoidable circumstance for groundwater transport in natural settings. The models have demonstrated an ability to simulate the tritium tracer elution to a reasonable degree of accuracy by using parameters derived, for the most part, from field data. Issues left outstanding include details of the cavity hydrology, the nature and extent of the low-conductivity layer, and the validity of a relatively large "K_d" for tritium (and ^{36}Cl).

In conclusion, the hydrologic modeling effort has been successful.

B. Noble-Gas Migration

The elution of ^{85}Kr appears to have been slightly delayed (~15%) relative to the elution of tritium – possibly as the result of retardation or an initial spatial distribution different from that of tritium.

C. Anion Migration and Exclusion

Four different anion-forming radionuclides (^{36}Cl, ^{129}I, ^{99}Tc, and ^{106}Ru) migrated the 91 m from RNM-1 to RNM-2S at a rate comparable to the rate of tritiated water movement (velocity of the advection front). In each case, there is reason to believe that the migrating species was, or could have been, an anion. To date, these are the only radionuclides other than tritium and ^{85}Kr that are known to have migrated to RNM-2S from RNM-1.

The delay of the tritium elution relative to the ^{36}Cl elution is more reasonably attributed to repulsion of the ^{36}Cl anion by the negative surface charge of minerals than to preferential sorption of HTO relative to H_2O. Positive identification of the source of this effect by experimental means, although feasible, is probably not important for predictive purposes.

D. Migration of Cationic Species

As mentioned above, Hoffman, Stone, and Dudley[2] reported that the concentrations of the radionuclides ^{90}Sr, ^{137}Cs, and ^{239}Pu were greatly reduced relative to those

of tritium in water samples from RNM-1. This reduction was attributed to a combination of incorporation into the glassy debris and sorption onto mineral surfaces.

It has been reported that none of these isotopes has been detected in water samples from RNM-2S. Such negative results are reassuring, but by themselves are not useful in assessing the environmental impact of testing. Ideally, the results must be related to source strength, dissolution, dilution, and migration so that a prediction can be made for the future distribution of the isotopes.

What has been learned about migration of these isotopes from the Cambric migration experiment? When considering the possibility for migration at Cambric, one can pose several questions regarding the isotopes that could, at least in principle, be answered by measurements at RNM-2S.

(1) Did the entire inventory of the isotope migrate freely with the same velocity as tritium?

(2) Did all, or some portion, of the isotope initially present in the water at RNM-1 migrate freely with same velocity as tritium?

(3) Did any of the isotope migrate and, if so, with what velocity?

The answers to these questions require analyses with sensitivity sufficient to detect the isotope at RNM-2S. **For the first question**, the sensitivity must be sufficient to measure the selected isotope at a concentration determined by (1) its ratio to tritium in the Cambric device, (2) the concentration of tritium at RNM-2S at its peak value, and (3) the decay factors from time zero to the time of the measurement. Thus, the sensitivities required for ^{90}Sr and ^{137}Cs are given by

$$S(^{90}Sr, 1) = 0.00166 \times 1.4 \times 10^{11} \times 0.87 = 2.0 \times 10^{8} \text{ atoms/m}\ell, \text{ and}$$

$$S(^{137}Cs, 1) = 0.00503 \times 1.4 \times 10^{11} \times 0.88 = 6.2 \times 10^{8} \text{ atoms/m}\ell.$$

For the second question, we substitute for the first item the atom ratios to tritium in RNM-1 water from Ref. 2 (1.3×10^{-6} and 3×10^{-7}, respectively), which leads to sensitivity requirements of

$$S(^{90}Sr, 2) = 1.6 \times 10^{5} \text{ atoms/m}\ell, \text{ and}$$

$$S(^{137}Cs, 2) = 3.7 \times 10^{4} \text{ atoms/m}\ell.$$

The values from Ref. 2 represent averages for the three cavity zone samples; if it is assumed that the chimney sample represents an equal volume of source water, then the ratios are somewhat higher (~60%) than the values quoted above. Recently reported values for the analytical sensitivities for ^{90}Sr are 5×10^{5} atoms /mℓ; for ^{137}Cs, the sensitivities are 1×10^{6} atoms/mℓ in the 2-ℓ samples and 1000 atoms/mℓ in the 200-ℓ samples.[13]

Based on these values, it is clear that if either ^{90}Sr or ^{137}Cs had migrated *en toto* with the same velocity as that of the tritiated water, they would have been observed. One can estimate that if as little as 0.5% of the total ^{90}Sr or

0.0003% of the total ^{137}Cs produced in the Cambric device had migrated unimpeded, it would have been detected.

The reported sensitivity is not adequate to determine whether or not the ^{90}Sr actually in solution at RNM-1 migrated unimpeded because even if all of it had migrated with the tritium, the concentration at RNM-2S would have been about a factor of 3 below the detection limit.

The ^{137}Cs in solution did not all migrate unimpeded; one can estimate that if 3% of the ^{137}Cs in solution at RNM-1 had migrated with the tritium, it would have been at the detection level in the 200-ℓ samples; 5% would have been easily detected. The sensitivity for ^{137}Cs in the 2-ℓ samples analyzed by radiochemical separation and purification is inadequate by a factor of 30 to answer this question.

The third question can be addressed by the data in Table VI. It appears that both ^{90}Sr and ^{137}Cs are migrating, as is indicated by the reduction in the concentration of these isotopes in water recovered from the chimney zone. It is not readily apparent how one could deduce their velocity from the available data, which indicate a decrease in concentration of at least 1 order of magnitude since pumping started. It can also be argued that the reduction could be the result of a slow sorption onto the solid matrix instead of migration.

If one speculates for a moment, it may be noted that the tritium concentration at RNM-1 had dropped by ~4 orders of magnitude by the time it peaked at RNM-2S. If this is extrapolated to ^{137}Cs, assuming that the slower rate of reduction is caused by equilibrium with sorbed isotope, which results in a slower migration rate, one might anticipate the peak to arrive at ~100 Mm^3.

It is possible to estimate the maximum K_d for any sorbing species that has reached RNM-2S at the present time. The K_d for tritium is taken as 0.0 (or 0.8, according to Burbey and Wheatcraft), and the peak location as 5 Mm^3 volume pumped. The total volume pumped as of 1991 is about four times larger, so a migrating species with only 25% of its atoms in solution at any given instant should just now be peaking at RNM-2S. For 30% porosity and a matrix density of 2.7, the K_d would be 0.5, which is a very small value when compared to typical measured values for cationic species. (If $K_d(T) = 0.8$, then the K_d for the migrating species would be ~3.) To elute a sorbing species with a K_d of 50 mℓ/g would require ~1500 yr.

E. Particulate Migration

There is not sufficient data available for particulates in the water at RNM-1 or RNM-2S to justify any conclusions regarding migration.

XVIII. THE FUTURE FOR CAMBRIC

What remains to be learned from the Cambric migration experiment? Can we hope to learn more about the migration behavior of ^{90}Sr, ^{137}Cs, or ^{239}Pu by continuing the pumping at RNM-2S? Are there other long-lived isotopes available for study?

What about particulates? These questions should be addressed as part of the planning process for Cambric. A few relevant comments follow here.

The hydrologic modeling effort has been successful and there is little point in pursuing it further unless more information becomes available.

Continued routine measurements of the migration of tritium, ^{36}Cl, ^{85}Kr, ^{99}Tc, ^{106}Ru, and ^{129}I seem unlikely to shed much additional light on their migration behavior. However, attempts to unravel several peculiarities (for example, the annual fluctuation in ^{85}Kr) may prove interesting, and there is still only minimal data for ^{99}Tc. Questions raised about the "sorption" of tritiated water should be addressed by sorption experiments in the laboratory.

As the total volume pumped from well RNM-2S increases, the effects of dilution and dispersion on the concentration of any migrating species also increase, which makes it more difficult, or perhaps impossible, to detect them. Perhaps it is time to do some model predictions, using the most favorable set of parameters, to find out under what circumstances (pumped volume and concentration) any new radionuclides from Cambric might be detected at RNM-2S. Care should be taken to see that the sensitivity of planned measurements of isotope concentrations will permit us to draw meaningful conclusions from the results. Particular attention should be paid to actinides because of their high health hazard rating and because there have been very few attempts to measure them to date.

It is well established that a large fraction of the Cambric radionuclides were incorporated in melted glass and that only small concentrations were present in solution for potential migration. The question of leaching from the glass could be answered by measuring the concentration in well RNM-1 (with the RNM-2S pump turned off) or by laboratory leaching experiments, rather than by trying to transport slowly leaching radionuclides to RNM-2S. It seems likely that with a leach rate of $\leq 10^{-4}$/yr it will impossible to detect even an unimpeded radionuclide at RNM-2S. If well RNM-1 were to be reopened to the cavity region, it would allow access to water in contact with the glass melt produced by the Cambric explosion. It would then be possible to observe the decrease in radionuclide concentration in the water as it is swept away by pumping RNM-2S. One could subsequently allow the radionuclides to be leached from the glass with RNM-2S shut off, and the cycle could be repeated if desired. Perhaps, with careful design of the experiment, one could learn about both leach rates and migration rates for ^{90}Sr, ^{137}Cs, ^{239}Pu, and ^{241}Am.

There is the possibility of drilling a hole closer to RNM-1 to try to intercept slowly migrating radionuclides. Unfortunately, the actual initial distribution of radionuclides at Cambric was only coarsely defined in a vertical direction and not at all in a horizontal direction. At locations closer to the cavity, one would be more subject to miscalculations of the actual size and shape of the cavity. Also, one would not have a "before migration" set of concentrations to compare with the "after migration" data.

Study of the role of particulates in groundwater transport could be pursued at Cambric. The concentration of particulates at RNM-2S seems to be rather low, but it may be possible to collect an amount that is sufficient for identifying their source and for use in laboratory transport experiments. Based on observations of particulate transport at other sites, it may be useful to collect large

amounts of particulates, first at RNM-1 and then at RNM-2S, for assay of ^{90}Sr, ^{137}Cs, ^{239}Pu, or ^{241}Am.

The Cambric Migration Experiment has reached a cross road. Many of the originally defined goals have been achieved. Measurements and modeling of the elution have been completed, and data are available beyond the peak of concentration for those radionuclides that can reasonably be expected to migrate and be detected at RNM-2S. This appears to be an appropriate point at which to develop a new or revised set of goals that are based on what has been learned and what remains to be learned.

REFERENCES

1. I. Y. Borg, R. Stone, H. B. Levy, L. D. Ramspott, "Information Pertinent to the Migration of Radionuclides in Ground Water at the Nevada Test Site, Part 1: Review and Analysis of Existing Information," University of California Radiation Laboratory report UCRL-52078 Pt.1 (May 1976).

2. D. C. Hoffman, R. Stone, and W. W. Dudley, Jr., "Radioactivity in the Underground Environment of the Cambric Nuclear Explosion at the Nevada Test Site," Los Alamos National Laboratory report LA-6877-MS (1977).

3. D. C. Hoffman, "Field Study of Radionuclide Migration," *in* "Radioactive Waste in Geological Storage," S. Fried Ed., ACS Symposium Series 100 (American Chemical Society, Washington, DC, 1979).

4. K. Wolfsberg, W. R. Daniels, D. C. Hoffman, and B. R. Erdal, "Migration of Radionuclides in Geologic Media - Laboratory and Field Studies," *in* "Effluent and Environmental Radiation Surveillance, ASTM STP 698," J. J. Kelly, Ed. (American Society for Testing and Materials, Philiadelphia, Pennsylvania, 1980), pp. 252-254.

5. D. C. Hoffman, W. R. Daniels, K. Wolfsberg, J. L. Thompson, R. S. Rundberg, S. L. Fraser, K. S. Daniels, "A Review of a Field Study of Radionuclide Migration from an Underground Nuclear Explosion at the Nevada Test Site," International Atomic Energy Agency report IAEA-CN-43/469 (1983).

6. M. L. Wheeler, H. S. Ng, and S. E. Logan, "Selection of a Simulation Model for Analysis of Radionuclide Migration at the RNM Experiment Site, Nevada Test Site, Nevada," Los Alamos Technical Associates report LATA-LSL-29-01 (January 1982).

7. T. J. Burbey and S. W. Wheatcraft, "Tritium and Chlorine-36 Migration from a Nuclear Explosion Cavity," US Department of Energy publication 45050, DOE/NV/10384-09 (April 1986).

8. W. R. Daniels, Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1979--September 30, 1980," Los Alamos National Laboratory report LA-8670-PR (February 1981).

9. W. R. Daniels, Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1980--September 30, 1981," Los Alamos National Laboratory report LA-9192-PR (February 1982).

10. W. R. Daniels, Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1981--September 30, 1982," Los Alamos National Laboratory report LA-9691-PR (May 1983).

11. W. R. Daniels and J. L. Thompson, Comps. and Eds., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1982--September 30, 1983," Los Alamos National Laboratory report LA-10121-PR (April 1984).

12. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1983--September 30, 1984," Los Alamos National Laboratory report LA-10372-PR (February 1985).

13. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1984--September 30, 1985," Los Alamos National Laboratory report LA-10644-PR (January 1986).

14. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1985--September 30, 1986," Los Alamos National Laboratory report LA-11081-PR (August 1987).

15. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1986--September 30, 1987," Los Alamos National Laboratory report LA-11223-PR (February 1988).

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

17. J. L. Thompson, Comp. and Ed., "Laboratory and Field Studies Related to the Hydrology/ Radionuclide Migration Project, October 1, 1988--September 30, 1989," Los Alamos National Laboratory report LA-11947-PR (November 1990).

18. A. E. Ogard, J. L. Thompson, R. S. Rundberg, K. Wolfsberg, P. W. Kubik, D. Elmore, and H. W. Bentley, "Migration of Chlorine-36 and Tritium from an Underground Nuclear Test," Radiochim. Acta **44/45**, 213-217 (1988).

19. M. Kahn and J. Kleinberg, "Radiochemistry of Iodine," National Academy of Science - National Research Council Nuclear Science Series report NAS-NS-3062 (September 1977).

20. D. G. Coles and L. D. Ramspott, "Migration of Ruthenium-106 in a Nevada Test Site Aquifer: Discrepancy Between Field and Laboratory Results," Science **215**, 1235 (1982).

21. R. W. Buddemeier, "Transport of Colloidal Contaminants in Groundwater: Radionuclide Migration at the Nevada Test Site," Applied Geochem. **3**, 535 (1988).

22. W. R. Penrose, W. L. Polzer, E. H. Essington, D. M. Nelson, and K. A. Orlandini, "Mobility of Plutonium and Americium Through a Shallow Aquifer in a Semiarid Region," Environ. Sci. Tech. **24**, 228 (1990).

23. D. R. Champ, J. L. Young, D. E. Robertson, K. H. Abel, "Chemical Speciation of Long-Lived Radionuclides in a Shallow Groundwater Flow System," Water Pollut. Res. J. Can. **19**, 35 (1984).

24. Jean-Pierre Sauty, "An Analysis of Hydrodispersive Transfer in Aquifers," Water Resour. Res. **16**, 145 (1980).

25. W. R. Daniels, Ed., "Laboratory and Field Studies Related to the Radionuclide Migration Project, October 1, 1981--September 30, 1982," Los Alamos National Laboratory report LA-9691-PR (May 1983). (Section B. "Transport of Tritium from the Cambric Cavity Region to RNM-2S: Modeling," B. J. Travis, H. E. Nuttall, S. W. Hodson, and R. S. Rundberg.)

26. Science Applications, Inc., "Tabulation of Waste Isolation Computer Models," Office of Nuclear Waste Isolation report ONWI-78, SAI/OR-749-2 (1979).

27. L. F. Konikow and J. D. Bredehoeft, "Computer Model of Two-Dimensional Solute Transport and Dispersion in Ground Water," *in* US Geological Survey Technical Water-Resource Investigations, Book 7, Ch. C2 (1978).

28. B. J. Travis, "TRACR3D: A Model of Flow and Transport in Porous/Fractured Media," Los Alamos National Laboratory report LA-9667-MS (April 1983).

29. "A Model for Calculating the Effects of Liquid Waste Disposal in Deep Saline Aquifers," US Geological Survey, Water Resources Investigation report 76-61 (1976). (Developed by INTERCOMP Resources Development and Engineering, Inc. Updated in 1978, 1979, and 1982.)

30. L. D. Ramspott and R. D. McArthur, "Results of the Exploratory Drill Hole Ue5n, Frenchman Flat, Nevada Test Site," Lawrence Livermore National Laboratory report UCID-17392 (1977).

31. G. W. Thomas and A. R. Swoboda, "Anion Exclusion Effects on Chloride Movement in Soils," Soil Sci. **110**, 163-166 (1970).

32. R. W. Buddemeier and Dana Isherwood, Comps., "Radionuclide Migration Project 1984 Progress Report," University of California Radiation Laboratory report UCRL-53628 (April 1985). (Section 4. "Cambric Plume Study," M. R. Ruggieri and R. W. Buddemeier, pp. 48-54.)

33. R. W. Buddemeier, Comp., "Hydrology and Radionuclide Migration Program 1985-1986 Progress Report," University of California Radiation Laboratory report UCRL-53779 (September 1988). (Section 3A. "Infiltration and Recharge (Cambric Event Site)," M. R. Ruggieri, Scott Tyler, and R. W. Buddemeier, pp. 19-23.)

34. K. Marsh, Comp., "Hydrology and Radionuclide Migration Program 1987 Progress Report," University of California Radiation Laboratory report UCRL-53779-87 (March 1991).

35. R. W. Buddemeier, Comp., "Hydrology and Radionuclide Migration Program 1985-1986 Progress Report," University of California Radiation Laboratory report UCRL-53779 (September 1988). (Section 3B. "Vegetation Water Budgets," M.R.Ruggieri, R. W. Buddemeier, and R. Jacobson, p. 23.)