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Laboratory and Field Studies Related to the Radionuclide Migration Project

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Compiled and Edited by
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LABORATORY AND FIELD STUDIES RELATED TO THE RADIONUCLIDE MIGRATION PROJECT

Annual Report

October 1, 1980 - September 30, 1981

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ABSTRACT

FY-1981 laboratory and field studies related to the Radionuclide Migration project are described. Results are presented for radiochemical analyses of water samples collected from the RNM-1 well and the RNM-2S satellite well at the Cambric site. Data are included for tritium, ^{85}Kr, ^{129}I, and ^{36}Cl. The maximum-concentration tritium peak appears to have arrived at RNM-2S near the end of FY-1981. Laboratory studies emphasize the sorptive behavior of alluvium and tuff and its dependence on mineralogy. Results from batch measurements and crushed-rock and whole-core column studies are presented.

I. INTRODUCTION

A field study of the distribution of radionuclides around an underground nuclear explosion cavity was initiated in 1974, about 9 yr after detonation. The goals of this Radionuclide Migration (RNM) Project were to determine the rates of migration underground in various media at the Nevada Test Site (NTS) and to determine the potential for movement both on and off the NTS of radioactivity from underground nuclear explosions. There was particular interest in possible contamination of water supplies. It was also envisioned that the study might provide data applicable to the underground disposal of radioactive waste.

This ongoing project is sponsored by the Nevada Operations Office of the Department of Energy with the participation of the Los Alamos National

Laboratory, the Lawrence Livermore National Laboratory, the United States Geological Survey, the Desert Research Institute, and appropriate support organizations.

The site of the 0.75-kt nuclear test Cambric, which was fired May 14, 1965, beneath the water table in tuffaceous alluvium, was chosen for initial studies. It was anticipated that eventually tests in other geologic media would be examined.

The field studies can be divided into two phases: 1) The Cambric cavity region was re-entered and a well (RNM-1) was completed in May 1974. Samples were taken to determine the radionuclide distribution between the solid material and water at the time the experiment was started. 2) Then, starting in October 1975, water was pumped from a satellite well (RNM-2S) located 91 m from the Cambric cavity so as to induce sufficient artificial gradient to draw water from the Cambric cavity and provide an opportunity for study of radionuclide migration under field conditions. Details of the early stages of the RNM Cambric experiment are given in Refs. 1 and 2.

The laboratory program has primarily involved sorption-desorption studies of NTS alluvium and tuff and leaching studies of nuclear test debris.

II. CAMBRIC STUDIES

A. Continuation of Tritium, ^{85}Kr, and Gamma-Spectral Analyses (W. R. Daniels, F. O. Lawrence, J. L. Thompson, R. D. Aguilar, M. R. Cisneros, S. Maestas, A. J. Mitchell, P. Q. Oliver, and M. A. Ott)

Pumping of water from the RNM-2S satellite well at the Cambric site has continued at the rate of ~600 gal. per minute, and semiweekly samples were analyzed for tritium. Detailed data through FY-1980 for tritium concentrations were summarized in Ref. 3 and are brought up-to-date in Table I and Figs. 1 and 2. The tritium (T) concentration has apparently peaked just below 3 nCi/mℓ (values at time of measurement). Discontinuities in the plots correspond to periods when the pump was inoperative.

One of the so-called "pressurized" water samples was taken each month from the hose bib at RNM-2S and analyzed for ^{85}Kr, HT, and HTO. Detailed data are presented in Table II and Figs. 3 and 4. Earlier results are given in Ref. 3. The atom ratio of ^{85}Kr to T remains at approximately one-third that calculated for Cambric, suggesting that krypton travels in the water

TABLE I

TRITIUM CONCENTRATION IN WATER PUMPED FROM SATELLITE WELL RNM-2S

Sample Number	Σ Gallons Pumped ($x\ 10^{-6}$)	Date	T^a (pCi/mℓ)
G-1227	1256.0	10/03/80	2941.6
G-1228	1258.9	10/06/80	2945.9
G-1232	1262.4	10/10/80	2946.3
G-1233	1265.7	10/14/80	2943.4
G-1236	1268.3	10/17/80	2951.6
G-1237	1271.1	10/20/80	2922.8
G-1241	1274.6	10/24/80	2946.9
G-1242	1277.2	10/27/80	2953.1
G-1245	1279.9	10/30/80	2937.8
G-1246	1283.2	11/03/80	2955.7
G-1250	1286.6	11/07/80	2953.3
G-1251	1289.4	11/10/80	2945.0
Pump off[b]			
G-1253	1294.6	11/19/80	2858.1
G-1254	1299.9	11/25/80	2854.5
G-1255	1308.2	12/04/80	2876.7
G-1256	1313.4	12/10/80	2867.2
G-1257	1318.8	12/16/80	2873.3
G-1258	1325.3	12/23/80	2853.9
G-1259	1331.6	12/30/80	2853.1
G-1260	1337.9	01/06/81	2860.1
G-1261	1344.2	01/13/81	2833.9
Pump off[b]			
G-1262	1350.3	02/10/81	2828.0
G-1265	1350.1	02/17/81	2826.6
G-1266	1362.4	02/24/81	2858.4
G-1267	1368.7	03/03/81	2836.2

TABLE I. (cont)

Sample Number	Σ Gallons Pumped ($x\ 10^{-6}$)	Date	T^a (pCi/mℓ)
G-1268	1375.0	03/10/81	2846.6
G-1269	1381.4	03/17/81	2863.5
G-1270	1387.5	03/24/81	2824.1
G-1271	1393.8	03/31/81	2801.5
G-1272	1401.1	04/08/81	2806.6
G-1273	1406.4	04/14/81	2788.7
G-1274	1413.6	04/22/81	2846.6
G-1275	1420.6	04/30/81	2835.9
G-1276	1425.0	05/05/81	2818.0
G-1277	1431.0	05/12/81	2805.0
G-1278	1437.0	05/19/81	2762.8
G-1279	1443.2	05/26/81	2767.7
G-1280	1450.8	06/04/81	2747.7
G-1281	1456.1	06/10/81	2758.4
G-1282	1462.0	06/17/81	2752.3
G-1283	1467.2	06/23/81	2724.4
G-1284	1474.1	07/01/81	2750.3
G-1285	1476.6	07/06/81	2736.6
G-1286	1488.0	07/17/81	2745.4
G-1287	1490.6	07/20/81	2712.2
G-1288	1496.6	07/27/81	2728.5
G-1289	1504.9	08/06/81	2713.9
G-1290	1508.5	08/10/81	2715.6
G-1291	1515.3	08/18/81	2699.5
G-1292	1522.3	08/26/81	2703.3
G-1293	1527.7	09/01/81	2700.7
G-1294	1533.3	09/08/81	2727.2
G-1295	1540.2	09/16/81	2685.0
G-1296	1547.2	09/24/81	2694.1
G-1297	1552.4	09/30/81	2670.0

TABLE I. (cont)

Sample Number	Σ Gallons Pumped ($\times 10^{-6}$)	Date	T[a] (pCi/mℓ)
G-1298	1557.7	10/06/81	2675.0
G-1299	1564.8	10/14/81	2619.2

[a]Standard deviations of counting data <0.1%.

[b]The pump at RNM-2S was off from November 13 to November 15, 1980, and from January 20 to February 10, 1981.

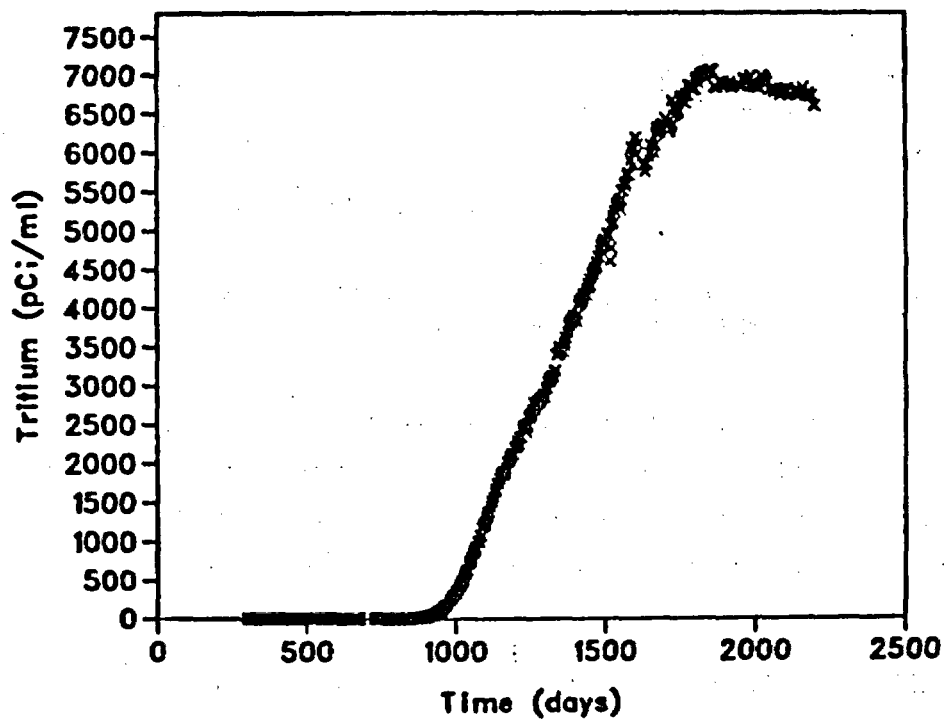

Fig. 1. Tritium concentration (corrected to Cambric zero time) in water pumped from RNM-2S vs time.

with the HTO but either at a slower rate or with some loss before or during transit. The ^{85}Kr concentration appears to have reached a maximum at about the same time as the tritium.

Starting in May 1978 a 55-gal. sample of RNM-2S water was taken each month in a plastic-lined barrel. Several additional samples were taken by adding the water to barrels already containing sufficient concentrated hydro-

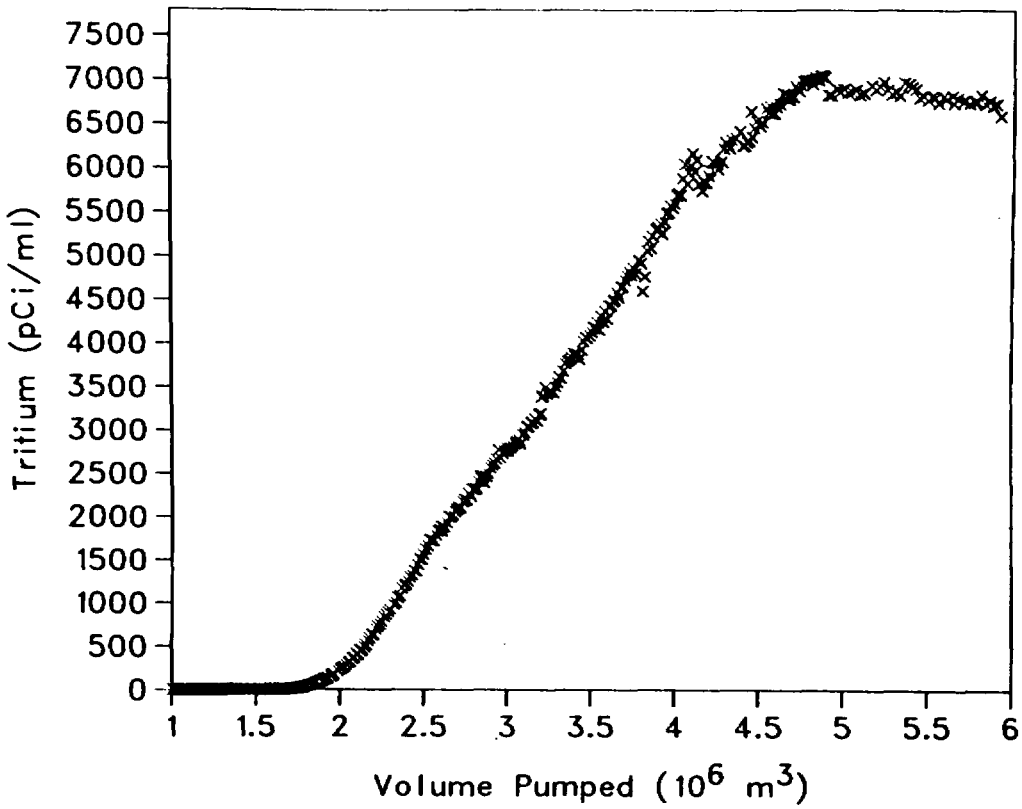

Fig. 2. Tritium concentration (corrected to Cambric zero time) in water pumped from RNM-2S vs volume pumped.

chloric acid to give a final concentration approximately 1 M in HCl. Twelve of the nonacidified samples have been concentrated to solids and encapsulated in plastic for gamma-spectral analysis.[3] Because of operational difficulties, results for the 55-gal. samples collected in FY-1981 are not yet available.

B. Transport of Tritium from Cambric Cavity Region to Well RNM-2S

(R. S. Rundberg)

The arrival of tritium at RNM-2S was compared with the calculations of Sauty[4] for an instantaneous tracer injection in a radial, converging flow field, similar to the RNM experiment. The shape of the elution curve depends on the Peclet number, which is inversely proportional to the dispersivity. The smaller the Peclet number the greater the dispersion, that is, the broader and more skewed the elution peak. To compare the experimental data with the calculation, dimensionless time t_r and dimensionless concentration C_r must be

TABLE II

ANALYSES OF PRESSURIZED WATER SAMPLES FROM RNM-2S[a]

Sample Number	I.D.	Date Collected	Total Volume from RNM-2S (10^6 m^3)	T Concentration (nCi/mℓ)	T Concentration (10^{11} atoms/mℓ)	^{85}Kr Concentration (dpm/mℓ)	^{85}Kr Concentration (10^6 atoms/mℓ)	$^{85}Kr/T$ (atom ratio x 10^8)	Ratio[b]
37	434-1-80-007	09/04/80	4.664	6.76	1.40	0.790	6.44	4.62	0.38
38	434-1-80-008	10/01/80	4.748	6.88	1.42	0.756	6.16	4.34	0.36
39	434-1-80-009	11/10/80	4.881	6.93	1.43	0.839	6.84	4.78	0.39
40	434-1-81-001	01/07/81	5.068	6.89	1.42	0.715	5.83	4.09	0.34
41	434-1-81-002	03/03/81	5.181	6.86	1.42	0.897	7.31	5.16	0.42
42	434-1-81-003	04/15/81	5.328	6.92	1.43	0.670	5.46	3.82	0.31
43	434-1-81-004	05/06/81	5.398	6.90	1.42	0.867	7.06	4.96	0.41
44	434-1-81-006	07/02/81	5.584	6.94	1.43	0.954	7.77	5.43	0.44
45	434-1-81-008	09/01/81	5.783	6.75	1.39	0.921	7.50	5.38	0.44
46	434-1-81-009	10/06/81	5.897	6.75	1.39	0.909	7.41	5.32	0.44

[a]All activity levels corrected to Cambric zero time (May 14, 1965).

[b]Ratio of measured to calculated $^{85}Kr/T$ atom ratio of 1.22×10^{-4} at Cambric zero time.

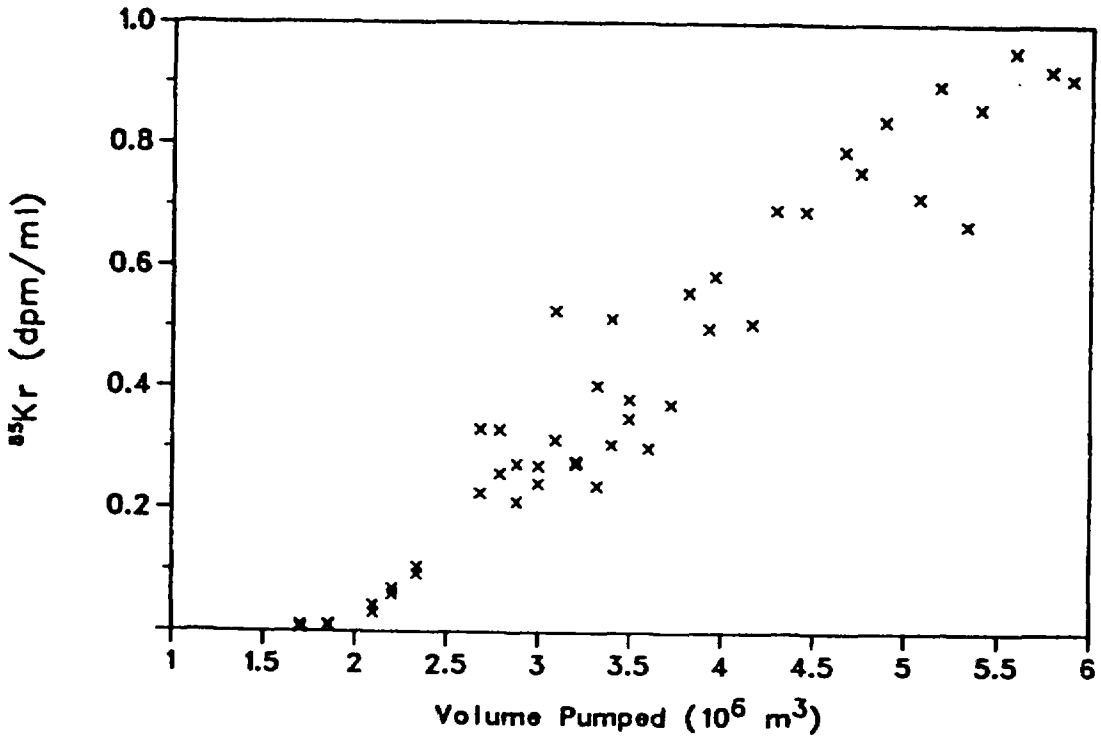

Fig. 3. The ^{85}Kr concentration (corrected to Cambric zero time) in water pumped from RNM-2S vs volume.

used. The dimensionless time is time t divided by the time when the maximum of the elution peak occurs; similarly, the dimensionless concentration is the concentration at time t divided by the maximum concentration.

Because the maximum in the tritium concentration in water from RNM-2S is not yet well defined, a calculation was made based on the amount of tritium pumped from RNM-2S by July 6, 1981, specifically 24% of that produced in Cambric. Values for t_r and C_r were determined for the same date (5.60×10^6 m^3 of water pumped) by locating the point on a calculated curve of Sauty corresponding to elution of 24% of the total tracer. The available[4] calculated curves for several Peclet numbers were thus compared with the normalized experimental data (Fig. 5). It appears that a calculated curve for a Peclet number between 3 and 10 (dispersivity between 30 and 10 m) would best fit the experimental data. This result can be compared with the data presented by Borg et al.,[5] who reported longitudinal dispersivity values that were estimated by calibrating mathematical models for transport against observed transport. Values ranged from 11.6 to 91 m for a wide variety of lithologies. The dispersivity for a sand

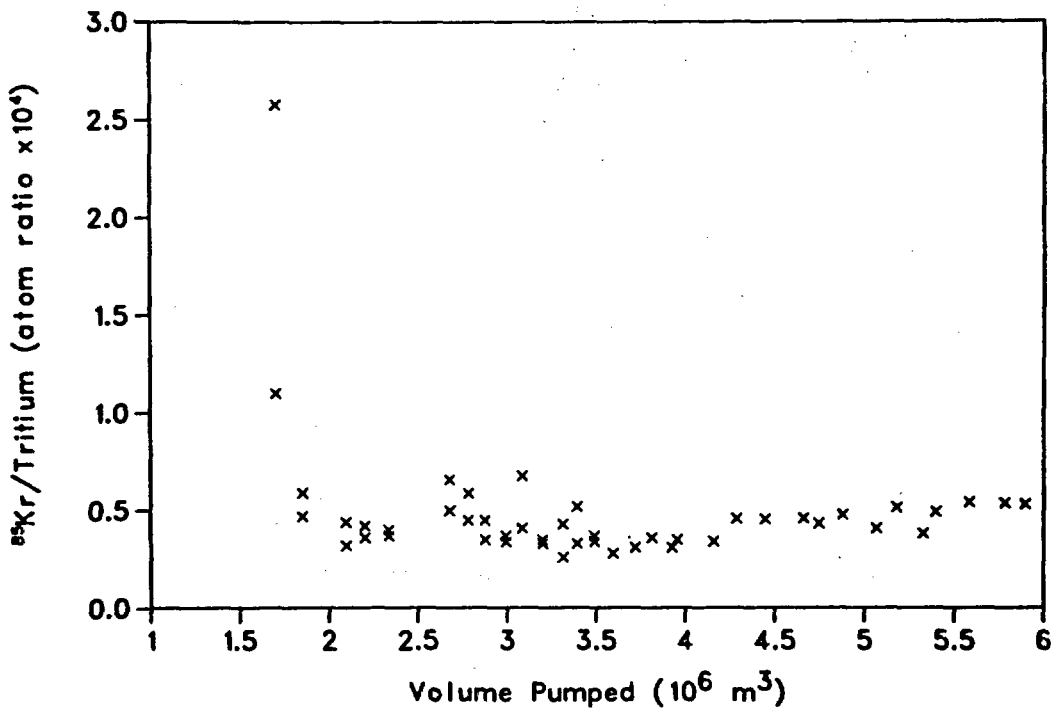

Fig. 4. Atom ratio of ^{85}Kr to tritium in water pumped from RNM-2S vs volume. (Values corrected to Cambric zero time.)

and gravel deposit, the lithology probably most closely resembling the tuffaceous alluvium of the current experiment, was found to be 21.3 m.

C. Determination of ^{36}Cl and ^{129}I in RNM Water Samples

The nuclide ^{36}Cl is produced in underground nuclear tests by neutron capture on stable ^{35}Cl (relative abundance 76%) that is in water, soil, and other materials in the vicinity of the tests. The ^{36}Cl/^{35}Cl ratios in two samples of water from the Cambric cavity have been measured by negative thermal-ionization mass spectrometry (TIMS) at Los Alamos to give values for the concentration of ^{36}Cl. A preliminary measurement of the ^{36}Cl/Cl ratio in one of the water samples by tandem-accelerator mass spectrometry (TAMS) gave a value in agreement with that by TIMS.

1. Thermal-Ionization Mass Spectrometry (K. Wolfsberg, P. L. Wanek, D. J. Rokop, and A. J. Gancarz). Silver chloride was precipitated from the groundwater without using any additional chloride carrier. The precipitate was dissolved in ammonium hydroxide, and the ammoniacal solution was dried on the side filaments of a triple filament assembly. Mass analyses of the Cl^- ions

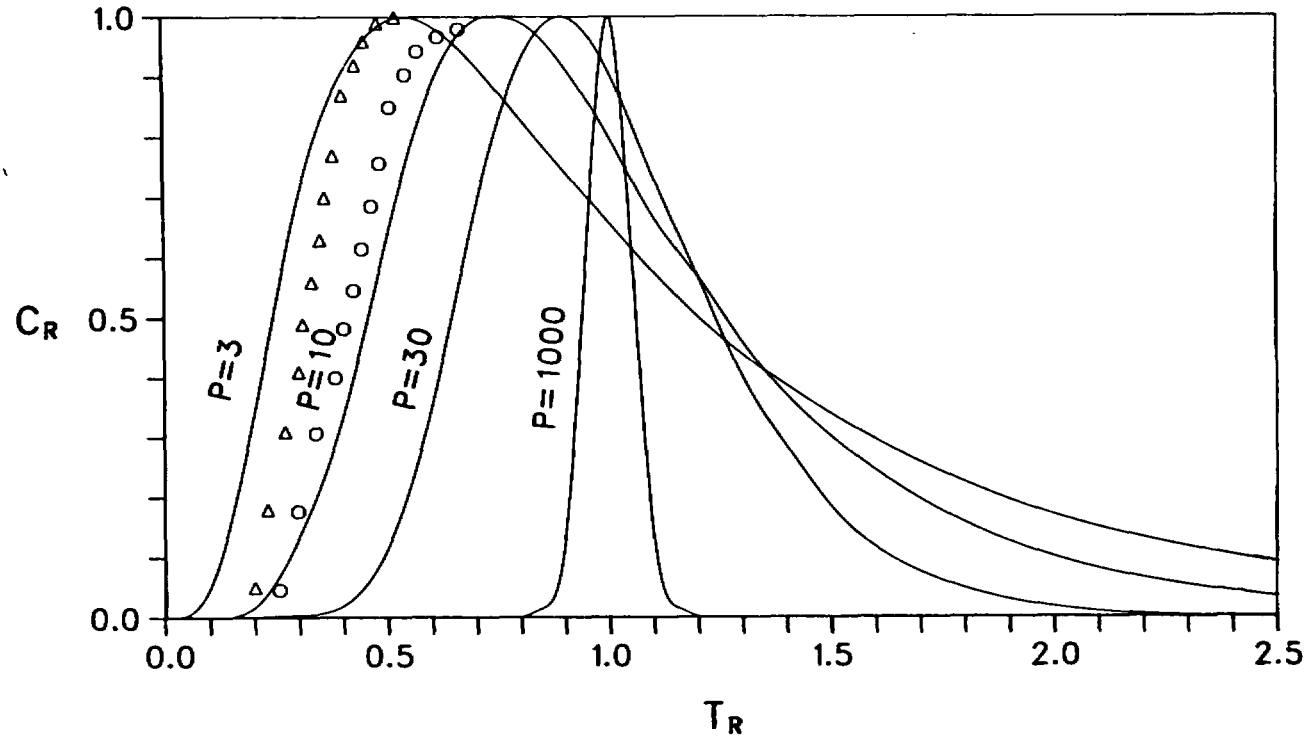

Fig. 5. Calculated elution of tracer for instantaneous tracer injection in a radial converging flow field for Peclet numbers 3, 10, 30, and 1000. Normalized data for tritium observed in RNM-2S water are shown by Δ for P = 3 and by O for P = 10.

were performed on both single magnetic sector and tandem magnetic sector thermal-ionization mass spectrometers.

Some chloride analyses were performed previously,[3] and others were being performed as part of this investigation by ion chromatography. The concentration of ^{36}Cl is then calculated from the chlorine isotope ratio and the chloride concentration. The results are given in Table III. The tritium in 1 mℓ each of water samples C30a and C66a represent ~1.4 x 10^{-10} of the total tritium from Cambric. If all of the ^{36}Cl produced is soluble and follows tritium, then a total of ~3 x 10^{21} atoms of ^{36}Cl would have been produced. We plan to measure additional $^{36}Cl/T$ ratios in waters from RNM-1 and RNM-2S.

Detection limits for the $^{36}Cl/^{35}Cl$ ratio are ~3 x 10^{-8} for the single-stage spectrometer and (1 to 5) x 10^{-9} for the tandem instrument. Improvements to ~1 x 10^{-10} seem possible. Thus, it should be possible to observe ^{36}Cl in the samples from RNM-2S having the highest T levels if the ^{36}Cl follows T.

TABLE III

CHLORINE-36 IN CAMBRIC WATERS

Sample	Mass Spectrometer	$^{36}Cl/^{35}Cl$	Natural Cl^- (mg/ℓ)	^{36}Cl (atoms/mℓ)
C30a[a]	single stage	2.81×10^{-7}	109[f]	3.94×10^{11}
C30a[a]	tandem	3.11×10^{-7}	109	4.36×10^{11}
C30a[a]	TAMS[b]			4.3×10^{11}
C30a[a]	TAMS[c]			6.3×10^{11}
C66a[a]	single stage	3.2×10^{-7}	109 ± 5[g]	4.49×10^{11}
G-3021[d]	single stage	$<4 \times 10^{-8}$	~20	
G-7496[e]	single stage	$<3 \times 10^{-8}$	~20	

[a]Descriptions of C series pumping from Cambric cavity are given in Ref. 1.

[b]Determined for sample of 0.5 mℓ of C30a water plus 300 mg of inactive chloride. Measured $^{36}Cl/Cl$ ratio was 4.2×10^{11} with estimated uncertainty of 10%.

[c]Determined on sample of 20 mℓ of C30a water plus 200 mg of inactive chloride. Measured $^{36}Cl/Cl$ ratio was 3.7×10^{9}, which was high enough to give problems in measurement. The number is not considered reliable.

[d]Composite of G-1272, -1273, and -1274 from RNM-2S at ~1.5×10^9 gal. pumped.

[e]Composite of samples G-74 to G-96 early in the pumping of RNM-2S.

[f]Not measured. Assumed to be the same as C66a. Data given in Ref. 1 indicate decreasing concentration with volume pumped.

[g]Measured by ion chromatography.

Exploratory experiments for measuring $^{129}I/^{127}I$ ratios are under way. For these experiments, it is necessary to dissolve precipitated AgI in NH_4CN. It appears that the limit of the $^{129}I/^{27}I$ ratio on the single-stage spectrometer is $\leq 5 \times 10^{-8}$. Because the concentration of natural iodine in the water is much less than that of natural chlorine, it will be possible to establish lower ^{129}I concentrations than ^{36}Cl concentrations with the TIMS method, assuming that the chemical separation problems can be solved.

2. Tandem-Accelerator Mass Spectrometry (K. Wolfsberg, P. L. Wanek, H. W. Bentley, D. Elmore, and L. E. Tubbs). We are exploring the use of TAMS techniques[6-8] for measuring lower concentrations of ^{36}Cl. It is possible to measure ^{36}Cl to total Cl ratios of 10^{-15} to 10^{-11} by the technique. For preliminary experiments, inactive chloride was added to samples of water from RNM-1. The amount added was much greater than that in the water, and ^{36}Cl concentrations are based on the added carrier. Silver chloride was precipitated. Additional purification of the samples from sulfur, which has isobaric interferences, was performed in Tucson, and the ratios were determined by TAMS at the Rochester Tandem Van de Graaff accelerator. The results of the analyses are given in Table III. The TAMS value obtained for the weaker (more inactive chloride) sample compares very favorably with those obtained for the same water by TIMS. The value for the stronger sample is still of the same order of magnitude, but the value is considered less reliable because it is outside of the range for analyses by TAMS and running of the sample resulted in contamination problems.

A series of samples with estimated $^{36}Cl/Cl$ ratios of 10^{-12} have been prepared for measurement early in 1982.

The TAMS technique can also be used to measure low $^{129}I/^{127}I$ ratios.[9] With the ability to measure such ratios in the 10^{-12} to 10^{-8} range, it is possible to add inactive iodide to water samples for purification and normalization. A series of iodide samples have been purified from the B, C, D, E, F, H, J, and K pumpings of RNM-1 and from selected times of the RNM-2S pumping. We hope to perform ^{129}I runs on these samples on an accelerator mass spectrometer in early 1982.

III. LABORATORY STUDIES

A. Sorption of Iodide, Ruthenium, Antimony, Tantalum, Iridium, and Arsenic on Alluvium (K. Wolfsberg and P. L. Wanek)

It was decided to perform another set of sorption experiments with alluvium using the nuclides leached from an underground test as tracers. The previous experiment of this type[10] gave sorption ratios for I^- in the range of 15 to 25 mℓ/g for sorption experiments and 400 to 900 mℓ/g for desorption experiments. These numbers do not seem reasonable now because we have since found that iodide does not appear to sorb at all ($R_d \cong 0$) on several different types of tuff.[11-13]

Alluvium from RNM-1 core 39 at a depth of 331.5 m was used in this experiment. The alluvium (not ground) was passed through a 500-μm sieve. Water collected between samples G-73 to G-97 from the RNM-2S well (pH ~8) was used. The alluvium samples were pretreated with the groundwater for 10 days to 3 wk before the experiment.

A combined sample of volatile and refractory debris from an underground explosion was shaken for several days with the water from the RNM-2S well. The water was separated from the solids by centrifuging and pipetting it off. The principal activities in the water at the beginning of this experiment are given in Table IV. Although the ^{131}I activity outweighed the others by orders of magnitude, it was possible to make quantitative measurements of ^{126}I, ^{103}Ru, ^{124}Sb, ^{182}Ta, ^{192}Ir, and ^{74}As in most of the samples. The activities of other nuclides were too weak for quantitative determination.

Detailed descriptions of sorption experiments are given in Refs. 11, 12, and 13. The results of the current sorption experiments (Table V) and desorption experiments (Table VI) are based on counting of aliquots of solids and of solutions after the experiments. Because a small amount of solution was entrapped in the solid material in each experiment, it was necessary to correct for the contributions of radionuclides in this aqueous phase to the observed activity in the solid.

Two of the sorption experiments (Table V) were performed with the supernates from the first set of sorption experiments rather than the solution described in Table IV. It was thought that if more than one species of an element is present, if only some of these sorb strongly, and if transformation from nonsorbing to sorbing species is slow compared to the times of our experiments (≤6 wk), then smaller R_d values would be observed in the second set of experiments. Within the experimental errors, the data in Table V indicate that this is not the case.

The sorption ratios for iodine, ruthenium, and antimony are significantly smaller than those reported in Ref. 10 for a different alluvium sample and a different starting leach solution. The low values for antimony are more in agreement with those values in Ref. 10 obtained with solutions of commercial ^{124}Sb tracer. Although the observed differences may be a result of differences in the solutions or alluvium, the earlier sample may have been contaminated with biological or organic material. Iodide is known to sorb on such materials (see, for example, Ref. 14).

TABLE IV

ACTIVITIES IN WATER AT BEGINNING OF EXPERIMENT

Nuclide	Activity (dis/min/10 mℓ)
^{131}I	1.0×10^{7}
^{126}I	2.2×10^{4}
^{103}Ru	2.0×10^{4}
^{124}Sb	1.1×10^{4}
^{126}Sb	1.8×10^{3}
^{74}As	3.8×10^{3}
^{182}Ta	5.8×10^{3}
^{192}Ir	3.4×10^{3}
^{196}Au	1.1×10^{4}

TABLE V

R_d VALUES (mℓ/g) FOR SORPTION ON AN ALLUVIUM SAMPLE

Contact Time (days)	Nuclide						
	^{131}I	^{126}I	^{103}Ru	^{124}Sb	^{182}Ta	^{192}Ir	^{74}As
7	0.2	0.2	10	1.5	8.2	2.3	5.0
14	0.3	0.1	7	1.8	6.1	1.7	4.1
28	"-0.2"		16	2.5	9.7	2.6	8.7
42	0.3		9	2.4	6.8	1.7	10[a]
18[b]			12	1.6	12.0	3.0	
40[b]			8	1.7	8.0		
Average	0.15 ± 0.17[c]		10 ± 3	1.9 ± 0.4	8.5 ± 2.1	2.3 ± 0.6	7.0 ± 2.8
Selected Average	0.2 ± 0.2		10 ± 3	2 ± 0.5	9 ± 3	2.3 ± 0.6	7 ± 3

[a]Fifty per cent uncertainty.

[b]Starting solutions for these runs were the supernatant solutions from the 28- and 42-day experiments above (see text).

[c]Standard deviation of the sample.

TABLE VI

R_d VALUES (mℓ/g) FOR DESORPTION

Contact Time (days)		Nuclide			
Sorption[a]	Desorption	^{131}I	^{103}Ru	^{124}Sb	^{182}Ta
7	21	6.4	300	10.9	93
14	14	2.9		10.1	33

[a]Traced alluvium sample from indicated sorption contact (Table V) used as starting material for desorption experiment.

Although the sorption ratio for ruthenium in this experiment is significantly less than the previously reported value, the number is nonetheless significantly greater than zero; the value for iodine may indeed be zero. Thus, one would expect that there would be significant retardation of ruthenium if migration occurs as chromatographic behavior, if conditions underground are as in the laboratory, and if the same species are involved.

B. Sorptive Behavior of Tuff

In conjunction with the Nevada Nuclear Waste Storage Investigations (NNWSI) program, the laboratory studies[3,13] of the partition of various radionuclides between NTS tuff and groundwater have been continued. Migration rate studies with columns of crushed rock and studies of flow and migration through whole and fractured cores were continued.

The distribution coefficient K_d is commonly used to describe the partition of a radionuclide between solid and aqueous phases at equilibrium. The term sorption ratio R_d, which does not imply equilibrium but is otherwise identical to K_d, is used here. The sorption ratio is defined as the ratio of the concentration of a species per gram of solid phase to the concentration of the species per milliliter of aqueous phase.

1. Sorption on Tuff Samples from the USW-G1 Drill Hole (S. D. Knight, A. J. Mitchell, D. L. Bish, K. Wolfsberg, and W. R. Daniels). Several types of batch experiments were performed with tuff samples from the USW-G1 drill hole at the NTS. The composition of the samples as determined by x-ray diffraction analyses is given in Table VII. Where two fraction sizes were used, data for each size are given.

One set of experiments involved the determination of sorption ratios for a series of pulverized tuffs with two size fraction, <75 μm and 75 to 500 μm, for each tuff. Sorption times were 3 and 6 wk, and corresponding desorption times were 6 and 3 wk. The averages of the R_d values for sorption and of the R_d values for desorption are given in Table VIII. There are generally no large differences in R_d values for sorption and desorption, with the value for desorption generally being slightly higher than that for sorption. For europium, the difference is greater, as has been observed before. The R_d values for sample G1-2476, which is not much different than the other devitrified tuffs, are significantly higher for desorption than for sorption.

Sorption ratios for sodium (^{22}Na), manganese (^{54}Mn), selenium (^{75}Se), and tin (^{113}Sn) were determined for four tuffs. The results for 2- and 4-wk sorption times and the corresponding 4- and 2-wk desorption times are given in Table IX. All experiments were done in air, except those with tuff G1-2237, which was done in a controlled atmosphere (nitrogen, $\leq$0.2 ppm oxygen, $\leq$20 ppm carbon dioxide).

Average values for sorption and desorption are given in Table X. As is expected for sodium, the R_d values are significantly higher for the two zeolitized samples (G1-2233 and G1-2289) than for the two devitrified ones (G1-2901 and G1-1883), and the values for desorption are greater than those for sorption. Values for selenium are low, and values for desorption are somewhat higher than those for sorption.

2. Dependence of Plutonium Sorption on Element Concentration

(M. R. Cisneros, W. R. Daniels, F. O. Lawrence, S. Maestas, and P. Q. Oliver). A batch technique was used to study the dependence of plutonium sorption on its concentration. Yields of traced feed solutions through the preparation procedure were variable but did not show any strong tendency to be lower at higher plutonium concentrations, as would be expected if solubility were the limiting factor. Tuffs from Yucca Mountain were used for these investigations: devitrified YM-22 and zeolitized YM-49. Results for both sorption and desorption experiments are given in Table XI.

A tendency toward higher R_d values at higher plutonium concentrations could be explained by precipitation, whereas lower R_d values could be the result of saturation of tuff sorption sites. However, within the accuracy of the data, no consistent trend can be observed although the R_d values for the highest plutonium concentrations and tuff YM-22 appear to be somewhat low.

TABLE VII

X-RAY DIFFRACTION ANALYSES OF TUFF SAMPLES

Sample	Particle Size (μm)	Composition (%)									
		Smectite	Illite/ Mica	Clino-ptilolite	Mordenite	Analcime	Quartz	Cristo-balite	Alkali Feldspar	Calcite	Glass
G1-1854	<75	5-10		40-60			20-40	15-30	10-30		
	75-500	5-10		30-50			5-15	15-30	20-40		
G1-1883		<2	<1				50-70	0-10	15-30		
G1-2233		<5	~5	20-40	20-40		15-30	10-20	10-20		
G1-2289			5-10	30-50	10-20		<5		30-50		
G1-2333	<75	5-10	<5				15-30	20-40	20-40		
	75-500	2-5	2-5				15-30	10-30	50-70		
G1-2410	<75	5-10	<5				20-40	5-15	30-50		
	75-500	5-10	<2				20-40	0-10	30-50		
G1-2476	<75	2-5	~2				30-50	5-15	40-60		
	75-500	<2	~2				30-50	5-15	40-60		
G1-2840	<75	2-5	2-5				40-60	0-10	30-50		
	75-500	2-5	2-5				40-60	0-10	30-50		
G1-2854	<75	<2	5-10				30-50	0-10	30-50		
	75-500	<2	5-10				30-50	0-10	30-50		
G1-2901		<2	<5				40-60	0-5	30-50	<5	

TABLE VIII

AVERAGE SORPTION RATIOS FOR CESIUM, STRONTIUM, BARIUM, AND EUROPIUM AND USW-G1 TUFFS[a]

Sample and Fraction Size (μm)	R_d (mℓ/g)							
	Cs		Sr		Ba		Eu	
	Sorption	Desorption	Sorption	Desorption	Sorption	Desorption	Sorption	Desorption
G1-2333, <75	1 600(100)	1 390(50)	218(2)	161(4)	1 860(40)	1 670(35)	2 600(600)	11 600(900)
G1-2333, 75-500	1 160(40)	1 070(50)	148(4)	119(7)	1 170(30)	1 270(170)	2 200(800)	8 300(1 500)
G1-2410, <75	2 020(20)	1 790(5)	280(4)	250(4)	3 040[b]	2 970(0)	420(30)	7 900(2 350)
G1-2410, 75-500	1 250(50)	1 120(100)	170(1)	140(15)	1 780[b]	1 750(150)	420(80)	6 000(3 000)
G1-2476, <75	870(50)	1 740(100)	50(1)	210(10)	500(20)	3 350(140)	4 800(300)	10 700(1 300)
G1-2476, 75-500	700(40)	1 530(5)	41(1)	202(4)	385(10)	3 700(65)	3 200(90)	15 200(0)
G1-2840, <75	2 640(160)	2 490(30)	170(1)	160(6)	2 500(200)	2 780(65)	5 600(600)	10 000(2 000)
G1-2840, 75-500	2 210(190)	2 040(40)	160(1)	134(1)	2 070(70)	2 280(15)	4 900(400)	7 960(70)
G1-2854, <75	1 100(600)	1 600(300)	90(30)	94(1)	4 100(2 000)	2 700(1 100)	1 800(800)	5 200(2 000)
G1-2854, 75-500	1 080(120)	1 160(20)	94(1)	97(1)	1 000(50)	1 330(0)	1 300(200)	5 000(200)
G1-1854, <75	15 000(100)	16 000(2 000)	81 000(11 000)	>70 000	56 000(8 000)	200 000(50 000)	>130 000	5 600(900)
G1-1854, 75-500	10 700(700)	13 000(2 000)	38 000(6 000)	73 000(31 000)	34 000(0)	110 000(30 000)	>120 000	3 900(900)

[a]Values in parentheses are the absolute-value standard deviations of the means.

[b]Only one measurement was available.

TABLE IX

SORPTION RATIOS FOR SODIUM, MANGANESE, SELENIUM, AND TIN AND USW-G1 TUFFS[a]

Sample	Contact Time (wk)		R_d (mℓ/g)			
	Sorption	Desorption	Na	Mn	Se	Sn
G1-2233[b]	2		148(3)	2 450(120)	14(7)	290(90)
	4		151(7)	630(80)	14(4)	140(30)
		2	151(18)	3 120(300)	130(70)	990(330)
		4	156(9)	1 520(120)	68(4)	500(20)
G1-2901	2		2.4(1.1)	>10 000	8.8(1.7)	>10 000
	4		4.1(0.4)	>10 000	8.7(4.7)	>10 000
		2	6.7(0.2)	>10 000	81(12)	>10 000
		4	5.5(0.0)	>10 000	66(3)	>10 000
G1-2289	2		224(7)	>10 000	16(7)	840(290)
	4		230(1)	6 650(620)	11(1)	600(50)
		2	267(6)	>10 000	53(5)	1 460(80)
		4	240(13)	>10 000	27(5)	1 030(20)
G1-1883	2		2.0(1.0)	128(8)	1.5[c]	232[c]
	4		2.6(1.1)	265(127)	10(6)	460(170)
		2	2.8(1.0)	560(390)	51(21)	2 540(120)
		4	2.7(1.4)	145[c]	40(20)	2 350(600)

[a]Sorption ratios are given as the average of two measurements. Numbers in parentheses are the absolute-value standard deviations of the means. Particle size range, <500 μm.

[b]Experiments performed under controlled-atmosphere conditions (nitrogen, ≤0.2 ppm oxygen, ≤20 ppm carbon dioxide).

[c]Only one measurement.

TABLE X

AVERAGE R_d VALUES FROM TABLE IX

	R_d (mℓ/g)[a]							
	Na		Mn		Se		Sn	
Sample	Sorption	Desorption	Sorption	Desorption	Sorption	Desorption	Sorption	Desorption
G1-2233[b]	150(4)	150(9)	1 500(500)	2 300(500)	14(3)	100(30)	210(60)	550[c](60)
G1-2901	2.9(0.6)	6.1(0.4)	>10 000	>10 000	8.8(2.0)	74(7)	>10 000	>10 000
G1-2289	230(3)	250(10)	>10 000	>10 000	14(3)	40(8)	720(140)	1 200(130)
G1-1883	2.3(0.6)	2.8(0.7)	132[c](6)	152[c](2)	6.9[c](4)	46(12)	380[c](120)	2 400(120)

[a]Sorption ratios are given as the average of four measurements, except where indicated. Numbers in parentheses are absolute-value standard deviations of the means.

[b]Experiments with tuff G1-2233 were performed under controlled-atmosphere conditions. Partical size range <500 μm.

[c]One value was rejected.

TABLE XI

DEPENDENCE OF PLUTONIUM SORPTION RATIO ON CONCENTRATION

Tuff Sample	Initial Plutonium Concentration[a] (M)		R_d (mℓ/g)			
			Sorption		Desorption	
	Added	Actual	<75 μm[b]	75-500 μm[b]	<75 μm	75-500 μm
YM-22	1.2×10^{-11}	8.0×10^{-12}	60	65	1 100	960
	5.6×10^{-11}	3.0×10^{-11}	63	62	1 200	580
	4.4×10^{-10}	2.1×10^{-10}	54	54	990	470
	3.9×10^{-9}	1.8×10^{-9}	66	41	1 100	920
	3.9×10^{-8}	1.0×10^{-8}	34	17	280	230
YM-49	1.2×10^{-11}	1.6×10^{-12}	170	150	700	670
	5.6×10^{-11}	7.7×10^{-12}	160	89	840	790
	4.4×10^{-10}	1.3×10^{-10}	130	140	780	590
	3.9×10^{-9}	4.3×10^{-10}	180	220	490	620
	5.4×10^{-8}	2.9×10^{-8}	270	240	690	340

[a]The plutonium concentrations at 100% yield based on assay of the ^{237}Pu and ^{239}Pu solutions are shown as "added." The plutonium concentrations actually present at the start of the batch contacts, given as "actual," are lower because of losses during preparation of the feed solutions.

[b]Particle size ranges were <75 μm and 75-500 μm.

3. Actinide Sorption Measurements Using Batch, Circulating System, and Crushed-Rock Column Methods (R. D. Aguilar, M. R. Cisneros, W. R. Daniels, F. O. Lawrence, A. J. Mitchell, and P. Q. Oliver). Experiments designed primarily to investigate the dependence of americium, plutonium, and neptunium sorption ratios on the method of determination were started. Batch, circulating system, and crushed-rock column methods are being studied. (The circulating system method is described in Refs. 13 and 15.) The original intent was to use a single traced feed solution for each nuclide and rock combination to eliminate possible variations in results with individually prepared batches of feed.

This was accomplished, except for the neptunium crushed-rock column experiments, which had to be repeated. Results to date for americium and neptunium sorption and desorption measurements by circulating system and batch methods are given in Tables XII and XIII. In most cases, sorption ratios obtained by the batch method are higher than those obtained with the circulating system, which is essentially a batch contact without physical movement of the rock. The difference in R_d values may be a result of high sorption on fine particles produced by self-grinding during the batch contacts. As usual, R_d values for desorption tend to be greater than those for sorption.[13] Tuff JA-37 from drill hole J-13 in Jackass Flats, which is high in montmorillonite, has the highest R_d values for americium. The relationship between sorptive behavior and mineralogy for these and other elements was discussed in Ref. 16.

Crushed-rock column experiments with neptunium and tuffs JA-37, YM-49, and G1-1883 are in progress. Preliminary results indicate that neptunium is retarded relative to tritiated water but that sorption ratios determined by the column method are somewhat lower than those from the batch method, as observed for other elements.[13]

Rock pretreatments and tracer preparations are in progress for neptunium batch sorption and desorption on a devitrified (YM-22), a zeolitized (YM-38), and a highly zeolitized (U12G-RNM) tuff.

4. Crushed-Rock Column Studies (E. N. Treher and N. A. Raybold). Elutions of radionuclides from ~40 columns of crushed rock have been completed or are in progress. The majority of these columns are of tuff although granite and argillite have also been studied to provide a comparison and to determine if general conclusions can be drawn that are applicable to all three rock types. Most of the columns have been loaded with ^{85}Sr, ^{137}Cs, and ^{133}Ba and run at flow rates of 13 to 67 m/yr. When elution peaks are observed, R_d values from these experiments are typically 1 to 3 times lower than R_d values from batch measurements on the same rock for ^{85}Sr and ^{137}Cs and 2 to 5 times lower for ^{133}Ba (Ref. 11). Some of the difference between results from column and batch measurements may be caused by fine material (<35 μm) present in many batch measurements and not present in the columns. The fine fractions are being examined for possible differences in mineralogy from the coarser fractions. Other explanations for the observed differences in R_d values are being sought.

TABLE XII

COMPARISON OF AVERAGE AMERICIUM SORPTION RATIOS[a] FROM CIRCULATING SYSTEM AND BATCH EXPERIMENTS

Tuff Sample	R_d (mℓ/g) Circulating System Sorption	Circulating System Desorption	Batch Sorption	Batch Desorption
YM-49	2 200(300)[b]	28 000(8 000)	4 300(1 400)	8 600(5 200)
JA-37	3 400(600)	110 000(40 000)	57 000(30 000)	52 000(4 800)
G1-1883	3 300(100)	36 000(7 000)	4 700(300)	7 000(900)
U12G-RNM9	3 100(700)		800(50)	

[a]Nonweighted averages of data from 3-, 6-, 9-, and 12-wk contact times, except U12G-RNM9, circulating, 3- and 6-wk, only. Rock particle size 106 to 250 μm.

[b]Values in parentheses are standard deviations of the means.

TABLE XIII

COMPARISON OF AVERAGE NEPTUNIUM SORPTION RATIOS[a] FROM CIRCULATING SYSTEM AND BATCH EXPERIMENTS

Tuff Sample	R_d (mℓ/g) Circulating System	Batch
YM-49	---	8.5(2)[b]
JA-37	---	29(7)
G1-1883	---	6(6)
U12G-RNM9	32(3)	31(9)

[a]Nonweighted averages of sorption data from 3-, 6-, 9-, and 12-wk contact times, except U12G-RNM9, circulating, 3- and 6-wk, only. Rock particle size 106 to 250 μm.

[b]Values in parentheses are standard deviations of the means.

A different kind of column behavior, a slow "leaking," is sometimes observed for cesium, particularly when clay/mica or zeolites are present in more than trace amounts. For these cases, the R_d value inferred from the position at which 50% of the activity has eluted from a column is usually greater than the values from the batch measurements on the same rock. The data from batch measurements and the mineralogic descriptions of these tuffs are summarized in Table XIV. For these samples, R_d values from desorption experiments are considerably higher than those for sorption, indicating, perhaps, that some portion of the ^{137}Cs may be irreversibly sorbed. Such an irreversible sorption might explain the slow leaking behavior observed from some columns.

Table XV shows the results from batch measurements and also mineralogic descriptions of tuffs for which R_d values from column experiments were less than those from batch experiments. For these rocks, all tuffs, the R_d values for sorption and desorption are quite similar, and the columns gave elution peaks.

TABLE XIV

DATA FOR SAMPLES FOR WHICH R_d VALUES FOR ^{137}Cs ARE GREATER FROM COLUMN MEASUREMENTS THAN FROM BATCH MEASUREMENTS

Column[a]	Description[b] (%)	Batch R_d (mℓ/g)	
		Sorption	Desorption
Granite [4]	5-15 m, <5 c, 40-60 AF, 30-50 Qz	320(30)	550(40)
Argillite [2]	10-20 m, 5-10 p, 20-40 sm, 15-30[c]	1 580(90)	2 680(140)
YM-38 tuff	5-15 sm, 50-70 cpt, 5-10 Qz, 5-10 AF, 10-20 cr	8 650(1 710)	13 100(0)
JA-37 tuff	20-40 sm, 5 i/m, ~5 cpt, 30-60 Qz, 15-30 AF	610(40)	850(50)

[a]The numbers in brackets indicate the number of columns completed to date.

[b]m = mica, c = chlorite, AF = alkali feldspar, Qz = quartz, p = pyrophyllite, sm = smectite, i/m = illite/mica, cpt = clinoptilolite, cr = cristobalite, tr = trace.

[c]The errors quoted in parentheses are the standard deviations of the means.

TABLE XV

DATA FOR SAMPLES FOR WHICH R_d VALUES FOR ^{137}Cs ARE LESS FROM COLUMN MEASUREMENTS THAN FROM BATCH MEASUREMENTS[a]

Column	Description (%)	Batch R_d (mℓ/g) Sorption	Batch R_d (mℓ/g) Desorption
YM-54(3)	tr mnt, tr i/m, 60 Qz, 30 AF	250(2)	310(20)
YM-22	<5 mnt, 40-60 Qz, 40-60 AF	340(60)	400(34)
1883A	<10 mnt, <5 m, 30-50 Qz, 50-70 AF	400(20)	430(3)

[a]See footnotes a, b, and c of Table XIV. mnt = montmorillonite.

Another unusual behavior with crushed-rock columns was observed during the elution of ^{85}Sr from a column of G1-1292 tuffs (Fig. 6). This sample is a vitrophyre containing 80 to 90% glass, 5 to 10% cristobalite, 10 to 20% alkali feldspar, and a trace of montmorillonite. The broad elution curve from this column is the only such curve yet seen for ^{85}Sr. Typically strontium elution peaks are sharp and fairly symmetrical as is shown in Fig. 7 for sample G1-1883, which contains primarily quartz and alkali feldspar. The point at which 50% of the ^{85}Sr was eluted from column G1-1292 was well past the peak (Fig. 6). In all other cases, the volume at which 50% of the ^{85}Sr eluted corresponded quite closely to the maximum of the curve.

5. Radionuclide Transport by Fracture Flow (J. L. Thompson). Several experimental techniques related to the transport of radionuclides by fluid flow in fractured rock are under development. It is anticipated that these techniques will aid in our efforts to model flow in fractured rock.[3] One technique involves use of a solution of an alpha emitting radionuclide to trace the fluid flow path through a fractured rock core. The other technique uses radionuclides to measure the diffusion coefficient for a dissolved species penetrating a rock matrix. The diffusion coefficient may be used to evaluate the pore constrictivity-pore tortuosity factor α/τ^2, which currently has to be estimated.

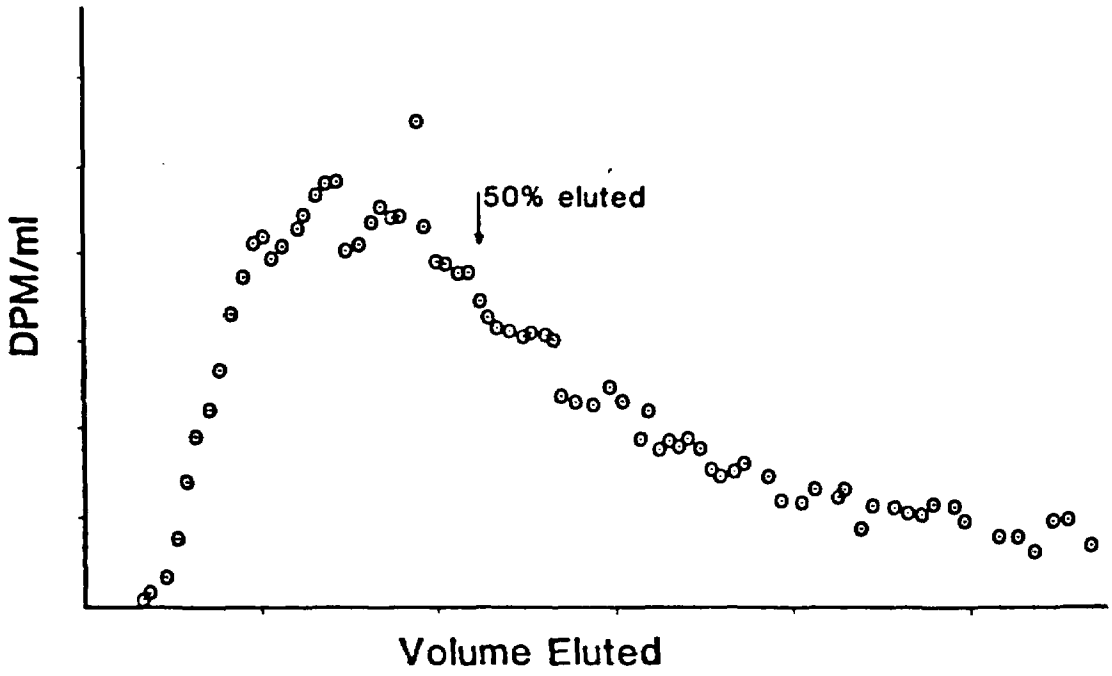

Fig. 6. Elution of ^{85}Sr from column G1-1292.

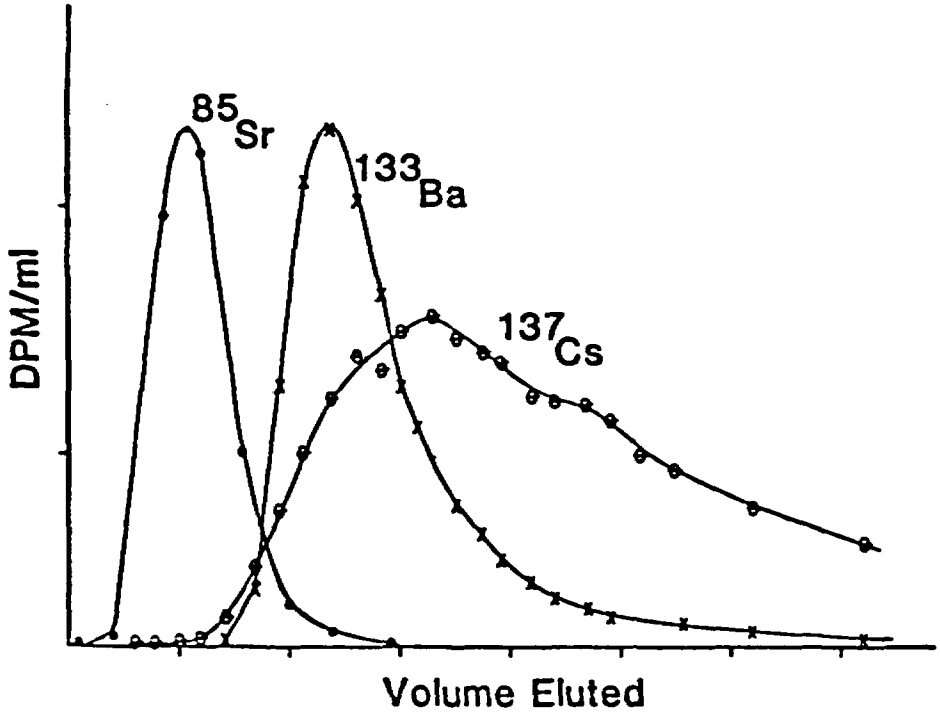

Fig. 7. Elution of nuclides from column G1-1883.

Cracked cores of Climax Stock granite were used in experiments directed toward developing a method of tracing fluid flow paths in laboratory experiments. Small cores, 2.54 cm in diameter and 1.6 cm in length, were placed in a system in which the pressure, and hence the crack aperture, could be controlled. A solution of ^{241}Am in synthetic groundwater was pumped into a crack, and the flow was stopped when the activity began to appear in the effluent. The levels of activity on the inner surfaces of the fracture indicated the path taken by the solution as it flowed through the aperture. Activity levels were measured using Kodak NTB-3 emulsion coated on a thin polymeric film sandwiched between the core faces for an appropriate exposure time. This technique seems to offer good resolution of activity levels on the rather uneven surfaces of a fracture. It does, however, require at least moderate activity levels in order to be applied in reasonable time spans. In our work, times of a few days were needed to expose the films sufficiently.

Experiments directed toward developing a method for measuring radionuclide diffusion coefficients were performed on 2.54-cm-diam by 0.93-cm-long cores of tuff from the USW-G1 hole. A core was sealed in a polycarbonate tube with epoxy, and a solution of J-13 water containing a radionuclide was added to the tube. The tube and core were then immersed in a large beaker of J-13 water so that the levels of liquid inside and outside the polycarbonate tube were the same. The concentrations of radionuclide in both liquid reservoirs were measured over a period of several weeks. The first experiment was with ^{233}U solution and G1-1883 tuff. The results indicated that the ^{233}U was sorbed on the rock to a sufficient extent to prevent calculation of a diffusion coefficient. A second experiment using ^{131}I as I^- ion and G1-2840 tuff gave a diffusion coefficient of the expected order of magnitude, 10^{-7} cm^2s^{-1}. Another experiment employing tritiated water is planned for the near future.

Additional data and calculations pertinent to the study of flow through fractured and porous rock are given in Refs. 12, 13, 15, and 17 through 20.

IV. REPORTS AND PUBLICATIONS

A number of publications and Los Alamos reports were issued that include, but are not limited to, data from the RNM program.

1. B. R. Erdal, B. P. Bayhurst, B. M. Crowe, W. R. Daniels, D. C. Hoffman, F. O. Lawrence, J. R. Smyth, J. L. Thompson, and K. Wolfsberg, "Laboratory Studies of Radionuclide Transport in Geologic Media," in Underground Disposal of Radioactive Wastes, Vol. II, IAEA-SM-243/37 (International Atomic Energy Agency, Vienna, 1980), p. 367.

2. J. R. Smyth, W. R. Daniels, S. J. DeVilliers, B. R. Erdal, F. O. Lawrence, M. L. Sykes, E. N. Vine, K. Wolfsberg, "Zeolitic Tuff as a Barrier to Radionuclide Migration," Geol. Soc. Amer. Abstr. Progr. 12, 525 (1980).

3. E. N. Vine, B. P. Bayhurst, W. R. Daniels, S. J. DeVilliers, B. R. Erdal, F. O. Lawrence, and K. Wolfsberg, "Radionuclide Transport and Retardation in Tuff," in Scientific Basis for Nuclear Waste Management, Vol. 3, J. G. Moore, Ed. (Plenum, New York, 1981), p. 483.

4. K. Wolfsberg, R. D. Aguilar, B. P. Bayhurst, W. R. Daniels, S. J. DeVilliers, B. R. Erdal, F. O. Lawrence, S. Maestas, A. J. Mitchell, P. Q. Oliver, N. A. Raybold, R. S. Rundberg, J. L. Thompson, and E. N. Vine, "Sorption-Desorption Studies on Tuff. III. A Continuation of Studies with Samples from Jackass Flats and Yucca Mounain, Nevada," Los Alamos National Laboratory report LA-8747-MS (May 1981).

5. K. Wolfsberg and B. R. Erdal, Eds., "Research and Development Related to the Nevada Nuclear Waste Storage Investigations, October 1 - December 31, 1980," Los Alamos National Laboratory report LA-8739-PR (April 1981).

6. B. R. Erdal, W. R. Daniels, and K. Wolfsberg, Eds., "Research and Development Related to the Nevada Nuclear Waste Storage Investigations, January 1 - March 31, 1981," Los Alamos National Laboratory report LA-8847-PR (July 1981).

7. B. R. Erdal, W. R. Daniels, D. T. Vaniman, and K. Wolfsberg, Eds., "Research and Development Related to the Nevada Nuclear Waste Storage Investigations, April 1 - June 30, 1981," Los Alamos National Laboratory report LA-8959-PR (October 1981).

8. W. R. Daniels, K. Wolfsberg, D. T. Vaniman, and B. R. Erdal, Eds., "Research and Development Related to the Nevada Nuclear Waste Storage Investigations, July 1 - September 30, 1981," Los Alamos National Laboratory report LA-9095-PR (in press, 1981).

9. W. R. Daniels, Ed., "Laboratory Studies of Radionuclide Distributions Between Selected Groundwaters and Geologic Media, October 1 - December 31, 1980," Los Alamos National Laboratory report LA-8691-PR (February 1981).

10. W. R. Daniels, Ed., "Laboratory Studies of Radionuclide Distributions Between Selected Groundwaters and Geologic Media, January 1 - March 31, 1981," Los Alamos National Laboratory report LA-8818-PR (June 1981).

11. W. R. Daniels and K. Wolfsberg, Eds., "Laboratory Studies of Radionuclide Distributions Between Selected Groundwaters and Geologic Media, April 1 - June 30, 1981," Los Alamos National Laboratory report LA-8952-PR (August 1981).

V. ORAL PRESENTATIONS

Several talks describing aspects of the RNM program were given during FY-1980, and an abstract has been submitted for an additional presentation in November.

1. E. N. Vine, B. P. Bayhurst, W. R. Daniels, S. J. DeVilliers, B. R. Erdal, F. O. Lawrence, and K. Wolfsberg, "Radionuclide Transport and Retardation in Tuff," International Symposium on the Scientific Basis for Nuclear Waste Management, Materials Research Society, Boston, Massachusetts, November 17-20, 1980.

2. J. R. Smyth, W. R. Daniels, S. J. DeVilliers, B. R. Erdal, F. O. Lawrence, M. L. Sykes, E. N. Vine, and K. Wolfsberg, "Zeolitic Tuff as a Barrier to Radionuclide Migration," Geological Society of America, Annual Meeting, Atlanta, Georgia, November 17-20, 1981.

3. W. R. Daniels, F. O. Lawrence, S. Maestas, and P. Q. Oliver, "Sorption of Am and Pu on Various Geologic Media under Reduced-Oxygen Conditions," American Chemical Society regional meeting, New Orleans, Louisiana, December 10-13, 1980.

4. R. S. Rundberg, S. Maestas, and J. L. Thompson, "Radionuclide Migration: Laboratory Experiments with Isolated Fractures," International Symposium on the Scientific Basis for Nuclear Waste Management, Materials Research Society, Boston, Massachusetts, November 16-19, 1981.

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