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ASSESSMENT OF THE POTENTIAL FOR RADIONUCLIDE MIGRATION FROM A NUCLEAR-EXPLOSION CAVITY

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ASSESSMENT OF THE TOTENTIAL FOR RADIONUCLIDE MIGRATION FROM A NUCLEAR EXPLOSION CAVITY Darleane C. Hoffman and William R. Daniels

Introduction

A field study of the distribution of radionuclides around an underground nuclear explosion cavity was initiated in 1974, about nine years after detonation of the nuclear test. 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 Georgical Survey, the Desert Research Institute, and appropriate support organizations

The site of the 0.75-kt nuclear test Cambric, which was fired beneath the water table in tuffaceous alluvium 294 m below the surface, was chosen for initial studies. It was anticipated that eventually tests in other geologic media would be examined.

Cambric was chosen for a number of reasons: The Cambric explosion cavity is within the NTS Area 5 water-supply aquifer. It was predicted that sufficient time had clapsed so that the cavity and chimney had filled with groundwater to the preshot static water level, 73 m above the detonation point. If so, radionuclines might be present in the water and constitute a potential source for migration. The Cambric detonation point is only 294 m below ground surface, and thus the re-entry diffing and sampling operations would be less difficult and expensive than for some of the more deeply buried tests. The site in Frenchman Flat is far enough from the areas of active nuclear testing so that damage or interruption of the re-entry and sampling operations from those activities would be unlikely. Sufficient tritium (³H or T) was present to provide an easily measurable tracer for water from the cavity region. The postshot debris also contained plutonium, uranium, and fission preducts whose concentrations in the rubble and groundwater from

the cavity and chimney regions could be measured and compared. A summary of the intensity of the radionuclide source term at the time of re-entry is given in Table I. The small yield was expected to have had little effect on the local hydrology. Further, it was judged that the alluvium constituted a good medium for hydrologic studies because it was more permeable than tuff and did not have large fissures or cracks through which the water might selectively flow.

The field studies can be divided into two phases: The Cambric cavity region was re-entered and a well (RNM-1) was completed to a depth of 370 m. Samples were taken to determine the radionuclide distribution between the solid material and water at the time the experiment was started. Water was then pumped from a nearby satellite well (RNM-2S) so as to induce an artificial gradient sufficient to draw water from the Cambric cavity and provide an opportunity for the study of radionuclide migration under field conditions. A schematic diagram of the placement of RNM-1 and RNM-2S is shown in Fig. 1.

Details of the early stages of the RNM Cambric experiment are given in Refs. 1 and 2.

Cambric Re-entry

Three types of samples were removed from the RNM-1 re-entry hole into the Cambric cavity: sidewall cores, pumped water, and water with contained gases. As drilling progressed, a total of 57 sidewall core samples was taken from RNM-1 from 34 m below the surface to 50 m below the original detonation point (Fig. 2). One core from each depth was placed immediately in a nitrogenflushed, gas-tight, stainless-steel container for subsequent analyses of ⁸⁵Kr, HT, HTO, and for gamma-spectral analyses. Other core samples were sealed in water-tight plastic bags for later gamma-spectral and radiochemical analyses to determine the concentrations of the various radionuclides present. The cores were also subjected to lithologic examination to determine the composition of the medium at different depths. Solid samples and water removed from the sidewall cores from the lower cavity region were analyzed ridrochemically for $^{90}\mathrm{Sr}_{*}$ $^{137}\mathrm{Cs}$, and $^{239}\mathrm{Pu}$, and effective distribution coefficients (ratio of the concentration in or on the solid to the concentration in the aqueous phase) were determined. These effective distribution coefficients are a measure of both reteation in the fused material and sorption. The radionuclides were found to be almost entirely incorporated in or on the solid material (Table 11).

After sidewall core sampling had been completed and the hole had been cleaned, casing with appropriately placed, inflatable, external packers was installed. The packers were used to minimize external water movement in the annular space between the casing and the wall. Beginning at the bottom, the water in five zones was sampled successively by isolating the zones with internal packers and perforating the casing (Fig. 3). Gas-tight water samples were taken at depth, and additional water samples were removed to the surface by a submersible pump. Representative activity levels of the radioauclides detected in water from each zone are given in Table III. Ten years after the test most of the radioactivity and the highest concentrations of all radionuclides were still found in the region of the original explosion cavity. No activity was found 50 m below the cavity. Measurements of HT and HTO removed from the cores and gas-tight water samples showed that more than 99.9% of the tritium was present as HTO. Although some 85 Kr and tritium were found in the fractured zone above the explosion region, they were concentrated in the cavity region. The measured 85Kr to T ratios for water from the explosion cavity zone were consistent with the relative amounts resulting from the Cambric test; the 85 Kr seemed to be dissolved in the water. No krypton was observed in water or solid material from cores taken above the water table. Water from the region of highest radioactivity at the bottom of the cavity contained only tritium and $^{90}\mathrm{Sr}$ at levels higher than the recommended concentration guides (CG) for drinking water in uncontrolled areas. 346

By comparing the measured ratio of each nuclide detected in the water to the tritium in the water with the calculated ratio for the Cambric source term, an effective overall retention factor, \mathbf{E}_3 , for each nuclide (ratio of the total activity in or on the solid to the total activity in the aqueous phase) was estimated. (This calculation assumes that all the radionuclides had been retained below the water table and that the fraction of a nuclide not in the water was in the solid.)

$$E_{d} = \frac{(\Lambda_{X}/\Lambda_{T})_{Cambric}}{(\Lambda_{X}/\Lambda_{T})_{water}} = \frac{(\Lambda_{X}/\Lambda_{T})_{cambric}}{(\Lambda_{X}/\Lambda_{T})_{water}} = \frac{(\Lambda_{X}/\Lambda_{T})_{cambric}}{(\Lambda_{X}/\Lambda_{T})_{water}}$$
Substituting
$$= \frac{\Lambda_{T}}{\Lambda_{T}} = \frac{(\Lambda_{X}/\Lambda_{T})_{cambric}}{(\Lambda_{X}/\Lambda_{T})_{cambric}} = \frac{(\Lambda_{X}/\Lambda_{T})_{cambric}}{(\Lambda_{X}/\Lambda_{T})_{water}} = \frac{(\Lambda_{X}/\Lambda_{T})_{cambrid}}{(\Lambda_{X}/\Lambda_{T})_{water}} = \frac{(\Lambda_{X}/\Lambda_{T})_{cambrid}$$

$$E_d \cong \frac{A_{X \text{ Cambric}}}{A_{X \text{ water}}} \cong \frac{A_{X \text{ solid}}}{A_{X \text{ water}}}$$
.

The nuclides ⁹³Sr, ¹⁰⁶Ru, ¹²⁵Sb, ¹³⁷Cs, ¹⁴⁷Pm, and ²³⁹Pu were all found to have high retention factors, indicating they are either retained in the fused debris or highly sorbed on the solid material, or both (Table IV).

Satellite Well

The satellite well RNM-2S was located 91 m f.om the Cambric explosion cavity. Pumping was begun in October, 1975 at a rate of about 1 m³/min; in October 1977 the rate was increased to about 2.3 m³/min. Significant amounts of tritiated water, signaling arrival of water from the Cambric cavity region, were finally detected about two years later after a total of about 1.44 million m³ of water had been pumped from the satellite well. After almost six years of pumping, the tritium concentration in the pumped water appears to have peaked, as shown in Fig. 4. Discontinuities in the plot correspond to periods during which the pump was not operating.

Since the observation of tritium in water from the salellite well, samples with contained gases have been taken by pumping from RNM-2S and from RMI-1. Results for such semples from the satellite well indicate the presence of 85 Kr, a gas which apparently was retained in the water during transit. The concentration of BSKr in water pumped from RNM-2S also appears to have reached a maximum value (Fig. 5). The 85 Kr/T atom ritios are shown in Fig. 6. It should be noted that these are relatively constant at a value of approximately 0.5×10^{-4} , considerably lower than that of 1.22×10^{-4} calculated for Cambric. So far the reason for this is not known; however, this result may very well be related to events occurring at RNM-1. The pump and packers in RNM-1 were left in a configuration such that pumping removes water from Zones IV and V (Fig. 3), which are just above the cavity region; most of the water production is believed to be from Zone IV. Data for water pumped from RNM-1 are presented in Table V. The concentrations of tritium and $^{85}\mathrm{Kr}$ in vater from RNM-1 have decreased by more than a factor of 100, presumably as a result of pumping at the satellite well; however, the 85Kr concentration is apparently decreasing more slowly than the tritium concentration. The 85 Kr/T ratios obtained for samples collected from RNM-1 since pumping began at the satellite well are consistently greater than the calculated ratio.

More than a dozen 55-gallon (0.21-m³) water samples, taken at intervals from RNM-2S since the first observation of tritium, have been reduced to solid residues by evaporation, and the gamma-ray spectra have been observed. With the possible exception of a very small amount of Ru (concentration <1% of that produced in Cambric), no gamma-emitting nuclides have been identified in these samples. Radiochemical analyses of other water samples for the beta-emitting nuclide Sr have also given negative results.

After removal of more than 5.6 million m^3 (over 1.5 billion gallons) of water from the satellite well, only tritium, which is present as HTO and chemically the same as the water, and 85 Kr, which seems to be dissolved in the water, have been positively identified in water from RNM-2S.

The arrival of tritium at the setellite well was compared with the calculations of Sauty for an instantaneous tracer injection in a radial, converging flow field, similar to the Cambric 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, i.e., the broader and more skewed the clution peak. In order to compare the experimental data with the calculation, dimensionless time, \mathbf{t}_r , and dimensionless concentration, \mathbf{C}_r , must be used. The dimensionless time is time t divided by the time when the maximum of the clution peak occurs; likewise, the dimensionless concentration is the concentration at t divided by the maximum concentration.

Since the maximum in the tritium concentration in water from RMM-2S is no yet known, a calculation was made based on the amount of tritium pumped from RMM-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 x 10⁶ m³ of water pumped) by locating the point on a calculated curve of Samty corresponding to clution of 24% of the total tracer. The available ⁷ calculated curves for several Peclet numbers were thus compared with the normalized experimental data (Fig. 7). It appears that a calculated curve for a Peclet number between three and ten (dispersivity between 30 and 10 m) would best fit the experimental data. This result can be compared with the data presented by horg et al., ⁸ who reported longitudinal dispersivity values which 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 and gravel deposit, the lithology probably most closely resembling the tuffaceous alluvium of the current experiment, was found to be 21.3 m.—1t

also appears that the tritium concentration in water from RNM-2S was essentially at its maximum value as of July 1981.

Laboratory Experiments

Laboratory experiments have primarily involved measurement of the partition of various radionuclides of interest between groundwaters and crushed rock samples of various geologic media ("batch" studies). Some experiments involving the leach rates for the removal of various radionuclides from nuclear test debris have also been performed. In general, the rates for removal of a radionuclide from such debris were found to be quite low (Table VI).

Results for the batch measurement of the sorption of a number of radionuclides on tuffaceous alluvium and bentonite are given in Table VII (Refs. 9 and 10). It is worth noting that the sorption properties of tuffaceous alluvium are comparable to those of bentonite, which is being considered for use as an engineered barrier because of its excellent sorptive properties. Sorptiondescrption equilibria are approached slowly, and results obtained for sorption measurements, where the tracer is initially present in the aqueous phase, and for desorption measurements, where the tracer is initially present with the solid phase, frequently differ. We therefore emphasize that "distribution coefficients" calculated from such data may represent nonequilibrium conditions. In addition, it should be noted that such values for the laboratory experiments refer only to sorption phenomena and do not include the effects of retention in fused debris as do the results given in Table 11. The "irreversible" sorption or much slower desorption may be due to speciation changes, diffusion into minerals, crystallization reactions on solids, or non-ionic sorption of celloids or precipitates. These possibilities require further investigation.

A comparison of the results from laboratory sorption experiments (Table VII) and calculated retention factors from Cambric water samples (Table IV) indicates that most of the radioactivity from the explosion is incorporated in the fused debris rather than sorbed. In the upper cavity and chimney regions, where sorption is presumably the dominant process, the laboratory results and retention factors from field data are comparable.

The sorptive properties of tuff have been extensively studied, 11-13 and it has been demonstrated that there are minerals in the varieties of tuff at the Nevada Test Site that exhibit excellent sorptive properties for cations.

Such tuffs should provide a natural barrier against radionuclide migration to the biosphere.

Most nuclides were found to sorb well on the geologic media studied; however, sorption of anionic species such as those of iodine, technetium, and uranium, which is frequently complexed by carbonate in the groundwater, is normally quite low for all geologic media (see Table VII and Refs. 11 and 12, for example). The chemistry of the actinides and lanthanides in groundwaters is complex and poorly understood. A significant research effort is necessary to achieve proper understanding of actinide and lanthanide chemistry in near-neutral solutions for adequate prediction of transport behavior in natural systems.

It took tritium, which is assumed to have a distribution coefficient of zero, over two years to reach RNM-2S. We estimate that more than 13 years would be required at current pumping rates for a hypothetical radionuclide with a distribution coefficient of only 1 mL/g to travel the 91 m between the explosion zone and RNM-2S. The laboratory experiments indicate that most elements have K_d values much greater than this (see Table VII) and would not be expected in water from the satellite well for many years. For example, strontium has a K_d of approximately 200 mL/g and would not reach RNM-2S for 1500 years. Non-equilibrium effects or the presence of colloidal or other non-sorbing species might allow some nuclides to move more rapidly than expected; therefore, monitoring for radionuclides in water pumped from RNM-2S is continuing.

Summary and Conclusions

The source term for radionuclides in the region of the Cambric nuclear explosion has been determined. Drillback cores were obtained and analyzed, and water was pumped from several vertical zones and analyzed. Most of the radioactivity produced in the test was found to be retained in the fused debris with only low concentrations in the water which had been in contact with the debris for nearly ten years. Most of the radioactivity and the highest specific activities of all radionuclides were found to be in the region of the original explosion cavity. No activity was found 50 m below the cavity. Water from the region of highest radioactivity at the bostom of the cavity contained only tritium and ⁹⁰Sr at levels higher than the recommended concentration guides for drinking water in uncontrolled areas.

During nearly six years (over 1.5 billion gallons of water) of pumping from a satellite well located 91 m from the Cambric cavity, only tritium, which is present as HTO and chemically the same as the water, and ⁸⁵Kr, which seems to be dissolved in the water, have been positively identified in water removed from this well, although there is some evidence for the possible migration of minute amounts of ¹⁰⁶Ru (less than 1% of that produced by the explosion). These results are consistent with laboratory studies which indicate that, in general, radionuclide sorption is sufficiently high to preclude the migration of such nuclides from the original cavity to the satellite well in the near future. Pumping and radioassay of water from the satellite well will be continued to investigate the possible arrival of non-sorbing species.

ACKNOWLEDGMENTS

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TABLE 1
CAMBRIC SOURCE TERM TEN YEARS AFTER DETONATION

Nuclide	Half i.ife (years)	Activity (Ci)		
3 _H	12.3	3.4 x 10 ⁴		
85 _{Kr}	10.7	4.4		
90 _{Sr}	29	34		
106 _{Ru}	1.0	2.8		
¹²⁵ Sb	2.8	3.2		
¹³⁷ Cs	30	99		
144 Ce	0.78	0.4		
147 _{Pm}	2.6	33		
155 _{Eu}	5.0	6.4		

TABLE II
EFFECTIVE DISTRIBUTION COEFFICIENTS
IN THE LOWER CAVITY REGION

	Distribution Coefficienc		
<u>Nuclide</u>	(ml/g)		
90 _{S r}	≅10 ⁴		
137 _{Cs}	>104		
239 _{Pu}	≊10 ⁸		

TABLE III

REPRESENTATIVE ACTIVITY LEVELS IN CAMBRIC
WATER SAMPLES TEN YFARS AFTER DETONATION

	Activity	Activity Level (dpm/m2)					
Zone	Level (µCi/m²)	85 Kr	90 Sr	106 _{Ru}	125 _{Sb}	137 _{Cs}	239 _{Pu}
below cavity	bg ^a	bg	þg	bg	bg	bg	bg
lower cavity	6.1	800	8	11	5	1.6	≤0.003
upper cavity	3.8	1200	5	4	2	1.4	≤0.003
chimney	0.084	70	5	nd ^b	nd	0.8	nd
adjacent to chimney	0.028	20	0.2	nd	nd	0.2	nd
cg ^c	2 x 10 ⁵		0.67	22.2	222	44.4	11.1

abg = no activity detectable above background levels.

TABLE IV
RETENTION FACTORS FROM CAMBRIC WATER SAMPLES

Zone	90 _{Sr}	105 _{Ru}	125 _{Sb}	13/ _{Cs}	147 _{Pm}	239 _{Pu}
below cavity	bg ^a	bg	уď	bg	bg	bg
lower cavity	2.1×10^{3}	1.0 x 10 ²	2.9×10^2	2.5 x 10 ⁴	≥10 ⁶	$\gtrsim 3.2 \times 10^7$
upper cavity	1.6 × 10 ³	1.9 × 10 ²	3.6 x 10 ²	1.8×10^4	nd	≥1.9 x 10 ⁷
chimney	3.9 x 10 ¹	ndb	r-d	6.6×10^{2}	nd	nd
adjacent to chimney	3.1×10^2	nd	nd	1.1 × 10 ³	nđ	nd

abg = no activity detectable above background levels.

bnd = not detected.

^CCG = recommended (Refs. 3 and 4) concentration guide applicable to water in uncontrolled areas.

 $^{^{}b}$ nd = not detected.

TABLE V
TRITIUM AND 85 Kr ACTIVITY LEVELS IN RNM-1
GAS-TIGHT WATER SAMPLES

		Volume of Water	T	85 _{Kr}	85 Kr/T
Entry	Date	$from RNM-2S(m^3)^b$	(nCi/m²)	(dpm/m2)	(atom ratio) ^C
original-Zone IV	8-8-75	0	150	70	1.8 x 10 ⁻⁴
original-Zone V	8-14-75	0	38	13	1.3×10^{-4}
re-entr <u>'</u> I ^d	10-4-77	1.17 x 10 ⁶	3.2	75	9.4×10^{-3}
re-entry II ^d	11-30-77	1.34 × 10 ⁶	2.0	6	1.3×10^{-3}
re-entry ill ^d	9-4-79	3.50 x 10 ⁶	0.26	0.57	8.8 x 10 ⁻⁴

All activity levels corrected to Cambric zero time.

 $^{^{}m b}$ Volume of water removed from RNM-2S by indicated date.

The calculated 65 Kr/T atom ratio for Cambric is 1.22 x 10^{-4} .

The packer between Zones IV and V was drilled out before pumping was started at RNM-2S. Water pumped for "re-entry" samples can enter from perforations in both Zones IV and V, but most of the water is believed to be from Zone IV.

TABLE VI LEACHING DATA FOR HIGH-GRADED REFRACTORY DEBRIS

	Fraction Leached						
	Ground		Chunks				
Test Location	U3km	U3k i	U7ap	U7ap			
Shaking Time (days)	23.6	23. ս	35.3	35.3"			
Nucl ide		· · · · · · · · · · · · · · · · · · ·					
54 _{Ma}			2.9×10^{-4}	9.0 x 10 ⁻⁵			
85 _{Co}			4.4×10^{-4}	9.6×10^{-5}			
887.	<8 x 10 ⁻⁵	<6 x 16 ⁻⁵	<8 x 10 ⁻⁴	\$2 x 10 ⁻⁴			
95 _{Zr}	<8 × 10 ⁻⁶ ,	$<2 \times 10^{-6}$	<4 x 10 ⁻⁵	<5 x 10 ⁻⁵			
106 _{Ru}	9.6 x 10 ⁻⁵	3.1 x 10 ⁻⁵	5.1 x 10 ⁻⁵	1.3 x 16 ⁻⁵			
124 _{Sb}	5.0 x 10 ⁻³	2.6 x 10 ⁻³	2.5 x 10 ⁻⁴	4.4 x 10 ⁻⁴			
126 _{Sb}	5.6 × 10 ⁻³	2.9 x 10 ⁻³					
) 29 _{Te} m	2.7×10^{-3}	1.8 x 10 ⁻³	9.6 x 10 ⁻³	1.5 x 10 ⁻³			
131	2.0×10^{-2}	1.4×10^{-2}	8.8 × 10 ⁻³	7.8 × 10 ⁻¹			
137 _{Cs}	<2 x 10 ³	<2 x 10 ⁻³					
140 _{B-1}	6.5 x 10 ⁻⁵	5.1 x 10 ⁻⁵	2.2 x 10 ⁻⁴	9.6 x 10 ⁻⁵			
141 _{Ce} -	<2 x 10 ⁻⁶	<1 × 10 ⁻⁶	54 x 10 ⁻⁶	<3 × 10 ⁻⁵			
181 _w	1.1 x 10 ⁻²	3.8×10^{-3}	1.2 x 10 ⁻³	3.5 x 10 ⁻⁴			
188 ₆	7.7 × 10 ⁻³	4.5 > 10-4					
182 _{Ta}	·3 × 10 ⁻⁴	*2 × 10 ⁻⁴					
237 _U	2.5×10^{-3}	1.2 x 10 ⁻³	6.1 x 10 ⁻⁴	∴6 x 10 ⁻⁴			
239,240 _{Pu}	50 × 10 ⁻⁶	55 x 10 ⁻⁰					

^{*}Solution and container were purged with argon before leaching.

TABLE VII

DISTRIBUTION COEFFICIENTS FOR
ALLUVIUM AND BENTONITE^a

Distribution Coefficient (m3/g) Rentonite
Description Alluvium Sorption Sorption Descrition 200 170 ti(v ') 60 6 7 50 Sı. Ł 80 Sb 30 220 Ho^b 20 Ip 220 180 1700 2500 Sr Rub ٠, die Nh 1900 3500 1000 2200 4000 4000 6000 3800 B.i 2200 8000 8000 1800 Cs 7000 1300 9000 21000 Co >20000 >2000 *500 >2000 Cr >6000 Eu >5000 >2000 >1400

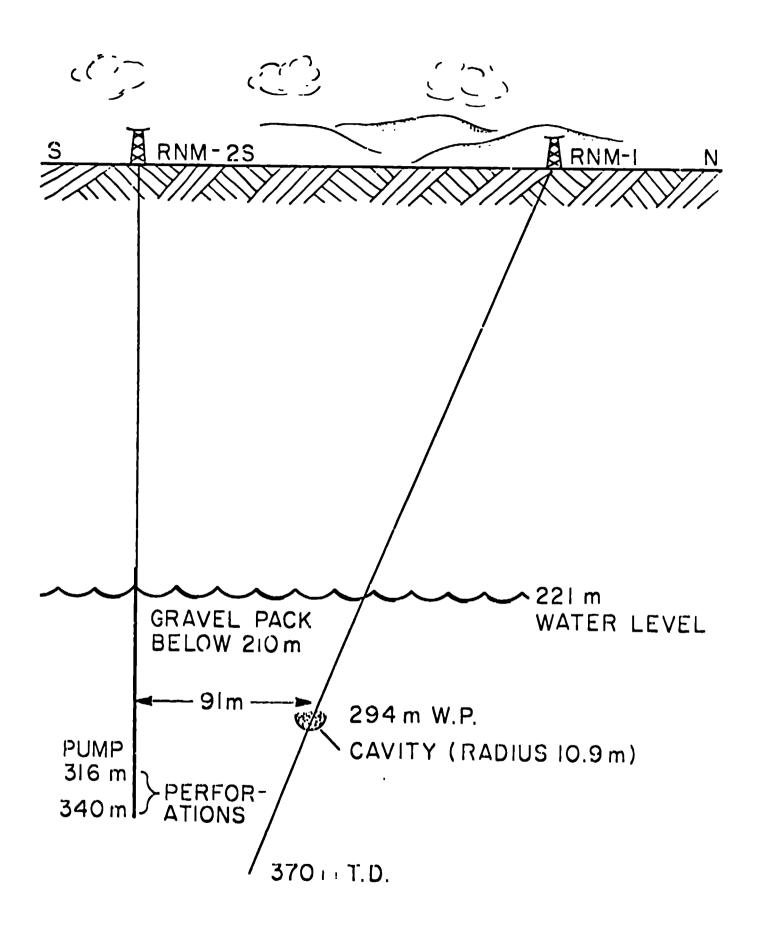
 $^{^{}m d}{
m Data}$ from Ref. 9, except 1 and Ru from Ref. 10.

^bTraced water produced by leaching test debris.

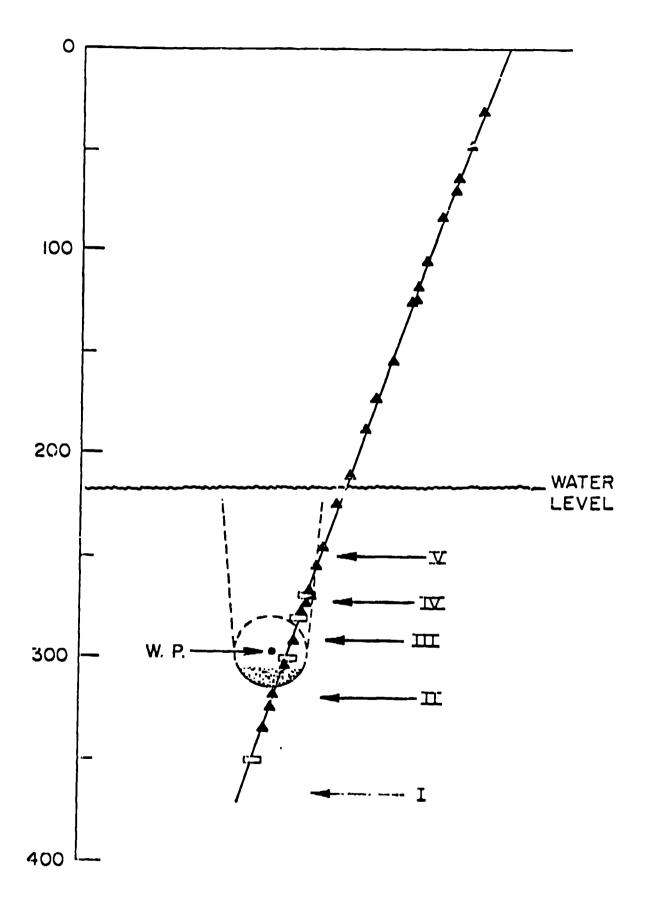
FIGURE CAPTIONS

- Figure 1. Schematic of RSM-1 and RNM-25.
- Figure 2. Locations of sidewall core samples taken from RNM-1 (Δ). Roman numerals indicate locations of isolated water sampling zones.

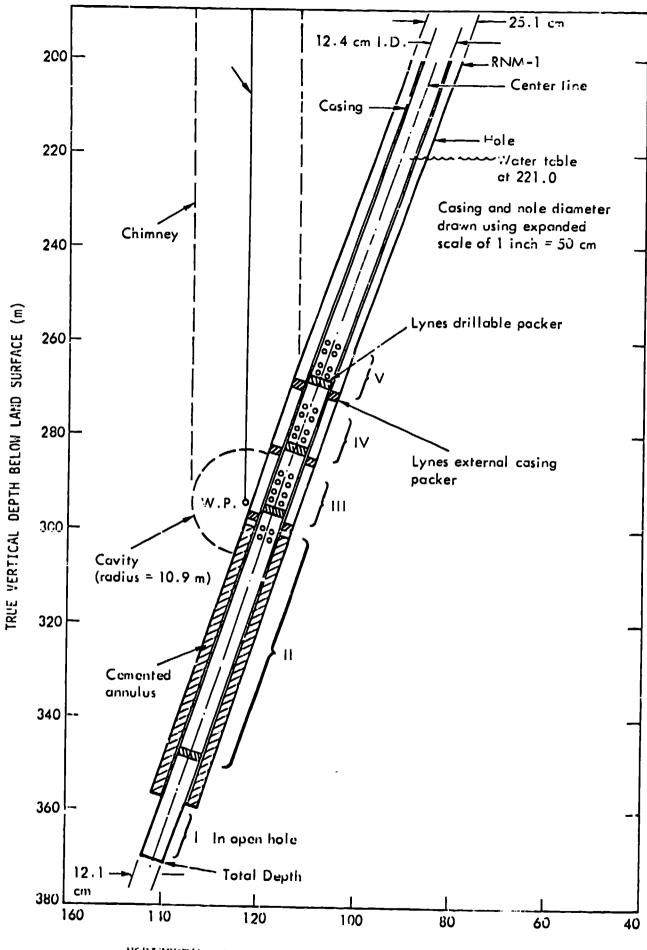
 The detonation point is indicated by W.P.
- Figure 3. Construction details of RNS-1 in and near the lower part of the Cambric chimney. Roman numerals indicate isolated water sampling zones. The original emplicement hole was designated U-be.
- Figure 4. Tritium concentration in water from RNM-2S, corrected to Cambric zero time.
- Figure 5. ** Kr concentration in water from RNM-28, corrected to Cambric zero time.
- Figure 6. 85 Kr/T atom ratios for water from RNM-2S. The calculated value for Cambric is 1.22×10^{-4} .
- Figure 7. Calculated elution of tracer for instantaneous tracer injection in a radial, converging flow field for bedlet numbers 3, 10, 30, and 1000 (from Ref. 6). Normalized data for tritium observed in RNM-28 water are shown by Δ for P = 3 and by O for P = 10.



Efgure 1



Fogure 2



HORIZONTAL DISTANCE SOUTH OF SURFACE SITE OF RHM-1 (m)

Pf page 3

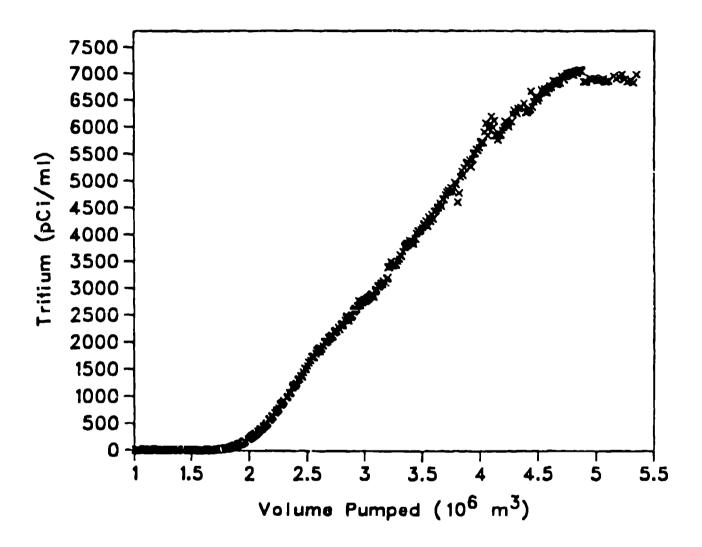


Figure 4

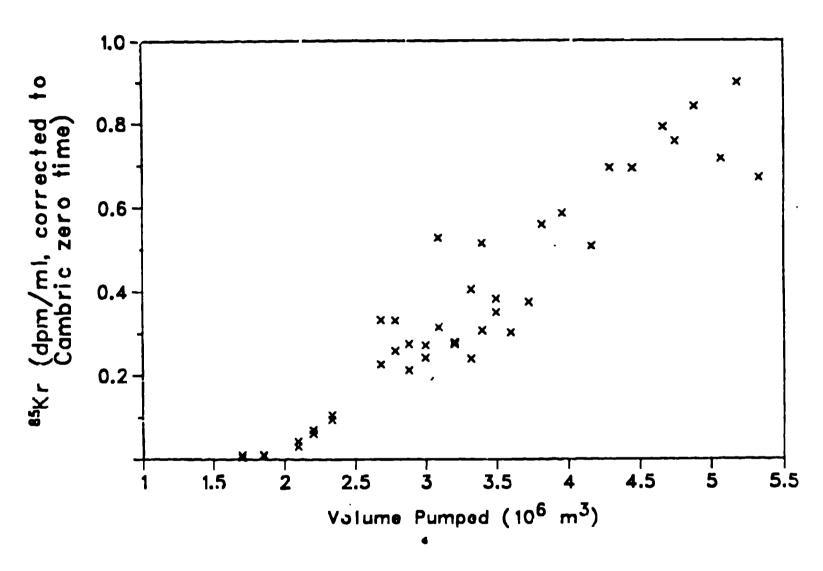


Figure 5

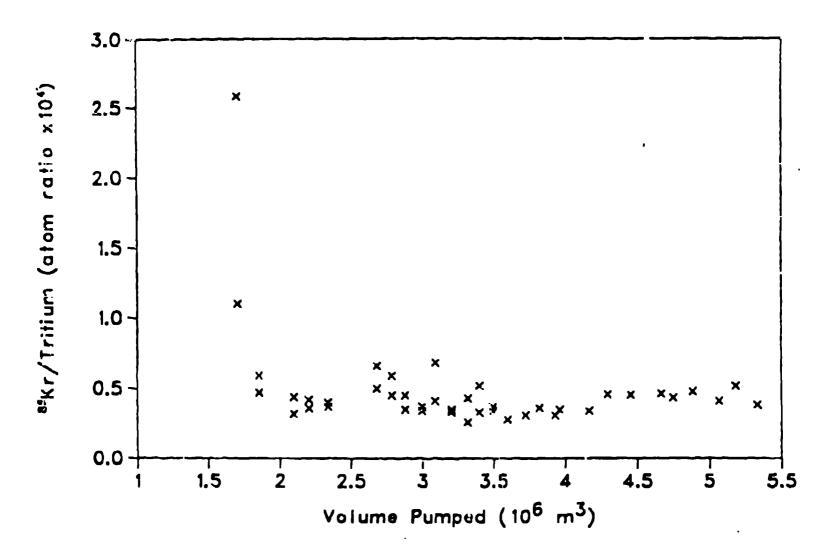


Figure 6

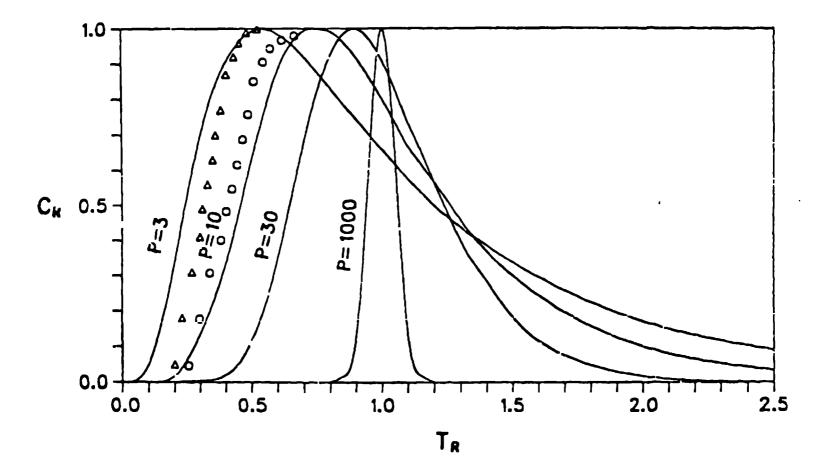


Figure 7