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ASSESSMENT OF THE POTENTIAL 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 Geological 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 elapsed so that the cavity and chimney had filled with groundwater to the preshot static water level, 73 m above the detonation point. If so, radionuclides 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 drilling 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 (^{3}H 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 products 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 nitrogen-flushed, gas-tight, stainless-steel container for subsequent analyses of ^{85}Kr, 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 radiochemically for ^{90}Sr, ^{137}Cs, and ^{239}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 retention in the fused material and sorption. The radionuclides were found to be almost entirely incorporated in or on the solid material(Table II).

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 radionuclides 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 ^{85}Kr 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}Sr at levels higher than the recommended concentration guides (CG) for drinking water in uncontrolled areas.[3,4]

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, K_d, 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.)

$$K_d = \frac{(A_X/A_T)_{\text{Cambric}}}{(A_X/A_T)_{\text{water}}} = \frac{(A_{X\ \text{Cambric}})(A_{T\ \text{water}})}{(A_{X\ \text{water}})(A_{T\ \text{Cambric}})}$$

Substituting $\frac{A_{T\ \text{water}}}{A_{T\ \text{Cambric}}} > 0.999 \sim 1$ (Ref. 1),

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

The nuclides ^{90}Sr, ^{106}Ru, ^{125}Sb, ^{137}Cs, ^{147}Pm, and ^{239}Pu 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 from the Cambric explosion cavity. Pumping was begun in October, 1975 at a rate of about 1 m^3/min; in October 1977 the rate was increased to about 2.3 m^3/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 m^3 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 satellite well, samples with contained gases have been taken by pumping from RNM-2S and from RNM-1. Results for such samples from the satellite well indicate the presence of ^{85}Kr, a gas which apparently was retained in the water during transit. The concentration of ^{85}Kr in water pumped from RNM-2S also appears to have reached a maximum value (Fig. 5). The $^{85}Kr/T$ atom ratios 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}Kr in water from RNM-1 have decreased by more than a factor of 100, presumably as a result of pumping at the satellite well; however, the ^{85}Kr 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-m^3) 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 ^{106}Ru (concentration <1% of that produced in Cambric),[5] no gamma-emitting nuclides have been identified in these samples. Radiochemical analyses of other water samples for the beta-emitting nuclide ^{90}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 satellite well was compared[6] with the calculations of Sauty[7] 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 elution peak. In order to compare the experimental data with the calculation, dimensionless time, t_r, and dimensionless concentration, C_r, must be used. The dimensionless time is time t divided by the time when the maximum of the elution 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 RNM-2S is not yet known, 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[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 Borg 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. It 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[9] 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. Sorption-descrption 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 II. 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 colloids 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 bottom of the cavity contained only tritium and ^{90}Sr 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 ^{85}Kr, 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 ^{106}Ru (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 I

CAMBRIC SOURCE TERM TEN YEARS AFTER DETONATION

Nuclide	Half Life (years)	Activity (Ci)
^{3}H	12.3	3.4×10^{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

Nuclide	Distribution Coefficient (mℓ/g)
^{90}Sr	$\cong 10^{4}$
^{137}Cs	$>10^{4}$
^{239}Pu	$\cong 10^{8}$

TABLE III

REPRESENTATIVE ACTIVITY LEVELS IN CAMBRIC WATER SAMPLES TEN YEARS AFTER DETONATION

Zone	Activity Level (μCi/mℓ)	Activity Level (dpm/mℓ)					
		^{85}Kr	^{90}Sr	^{106}Ru	^{125}Sb	^{137}Cs	^{239}Pu
below cavity	bg[a]	bg	bg	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]	2×10^5	---	0.67	22.2	222	44.4	11.1

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[a] bg = no activity detectable above background levels.

[b] nd = not detected.

[c] CG = recommended (Refs. 3 and 4) concentration guide applicable to water in uncontrolled areas.

TABLE IV

RETENTION FACTORS FROM CAMBRIC WATER SAMPLES

Zone	^{90}Sr	^{106}Ru	^{125}Sb	^{137}Cs	^{147}Pm	^{239}Pu
below cavity	bg[a]	bg	bg	bg	bg	bg
lower cavity	2.1×10^3	1.0×10^2	2.9×10^2	2.5×10^4	$\geq 10^6$	$\geq 3.2 \times 10^7$
upper cavity	1.6×10^3	1.9×10^2	3.6×10^2	1.8×10^4	nd	$\geq 1.9 \times 10^7$
chimney	3.9×10^1	nd[b]	nd	6.6×10^2	nd	nd
adjacent to chimney	3.1×10^2	nd	nd	1.1×10^3	nd	nd

---

[a] bg = no activity detectable above background levels.

[b] nd = not detected.

TABLE V

TRITIUM AND ^{85}Kr ACTIVITY LEVELS[a] IN RNM-1 GAS-TIGHT WATER SAMPLES

Entry	Date	Volume of Water from RNM-2S(m^3)[b]	T (nCi/mℓ)	^{85}Kr (dpm/mℓ)	^{85}Kr/T (atom ratio)[c]
original-Zone IV	8-8-75	0	150	70	1.8×10^{-4}
original-Zone V	8-14-75	0	38	13	1.3×10^{-4}
re-entry I[d]	10-4-77	1.17×10^{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 III[d]	9-4-79	3.50×10^{6}	0.26	0.57	8.8×10^{-4}

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[a] All activity levels corrected to Cambric zero time.

[b] Volume of water removed from RNM-2S by indicated date.

[c] The calculated ^{85}Kr/T atom ratio for Cambric is 1.22×10^{-4}.

[d] 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 Material		Chunks	
Test Location	U3km	U3ki	U7ap	U7ap
Shaking Time (days)	23.6	23.6	35.3	35.3[a]
Nuclide				
^{54}Mn			2.9×10^{-4}	9.0×10^{-5}
^{85}Co			4.4×10^{-4}	9.6×10^{-5}
^{88}Y	$<8 \times 10^{-5}$	$<6 \times 10^{-5}$	$<8 \times 10^{-4}$	$<2 \times 10^{-4}$
^{95}Zr	$<8 \times 10^{-6}$	$<2 \times 10^{-6}$	$<4 \times 10^{-5}$	$<5 \times 10^{-5}$
^{106}Ru	9.6×10^{-5}	3.2×10^{-5}	5.2×10^{-5}	1.3×10^{-5}
^{124}Sb	5.0×10^{-3}	2.6×10^{-3}	2.5×10^{-4}	4.4×10^{-4}
^{126}Sb	5.6×10^{-3}	2.9×10^{-3}		
$^{129}Te^{m}$	2.7×10^{-3}	1.8×10^{-3}	9.6×10^{-3}	1.5×10^{-3}
^{131}I	2.0×10^{-2}	1.4×10^{-2}	8.8×10^{-3}	7.8×10^{-3}
^{137}Cs	$<2 \times 10^{-3}$	$<2 \times 10^{-3}$		
^{140}Ba	6.5×10^{-5}	5.1×10^{-5}	2.2×10^{-4}	9.6×10^{-5}
^{141}Ce	$<2 \times 10^{-6}$	$<1 \times 10^{-6}$	$<4 \times 10^{-6}$	$<3 \times 10^{-5}$
^{181}W	1.1×10^{-2}	3.8×10^{-3}	1.2×10^{-3}	3.5×10^{-4}
^{188}W	7.7×10^{-1}	4.5×10^{-1}		
^{182}Ta	$<1 \times 10^{-4}$	$<2 \times 10^{-4}$		
^{237}U	2.5×10^{-3}	1.2×10^{-3}	6.1×10^{-4}	2.6×10^{-4}
$^{239,240}Pu$	$<6 \times 10^{-6}$	$<5 \times 10^{-6}$		

[a] Solution and container were purged with argon before leaching.

TABLE VII

DISTRIBUTION COEFFICIENTS FOR ALLUVIUM AND BENTONITE[a]

	Distribution Coefficient (mℓ/g)			
	Alluvium		Bentonite	
	Sorption	Desorption	Sorption	Desorption
U(VI)	6	60	200	170
Sn	6	80	7	50
Sb[b]	30	220		
Mo[b]	20			
I[b]	[illegible]	[illegible]	[illegible]	
Sr	220	180	1700	2560
Ru[b]	[illegible]	[illegible]	[illegible]	
Nb	1900	3500	1000	2200
Ba	3800	4000	4000	6000
Cs	8000	8000	1800	2200
Co	9000	21000	1300	7000
Ce	>20000	>2000	>500	>2000
Eu	>5000	>2000	>1400	>6000

[a] Data from Ref. 9, except I and Ru from Ref. 10.

[b] Traced water produced by leaching test debris.

FIGURE CAPTIONS

Figure 1. Schematic of RNM-1 and RNM-2S.

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 RNM-1 in and near the lower part of the Cambric chimney. Roman numerals indicate isolated water sampling zones. The original emplacement hole was designated U-5e.

Figure 4. Tritium concentration in water from RNM-2S, corrected to Cambric zero time.

Figure 5. ^{85}Kr concentration in water from RNM-2S, 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 Peclet numbers 3, 10, 30, and 1000 (from Ref. 6). Normalized data for tritium observed in RNM-2S water are shown by Δ for P = 3 and by O for P = 10.

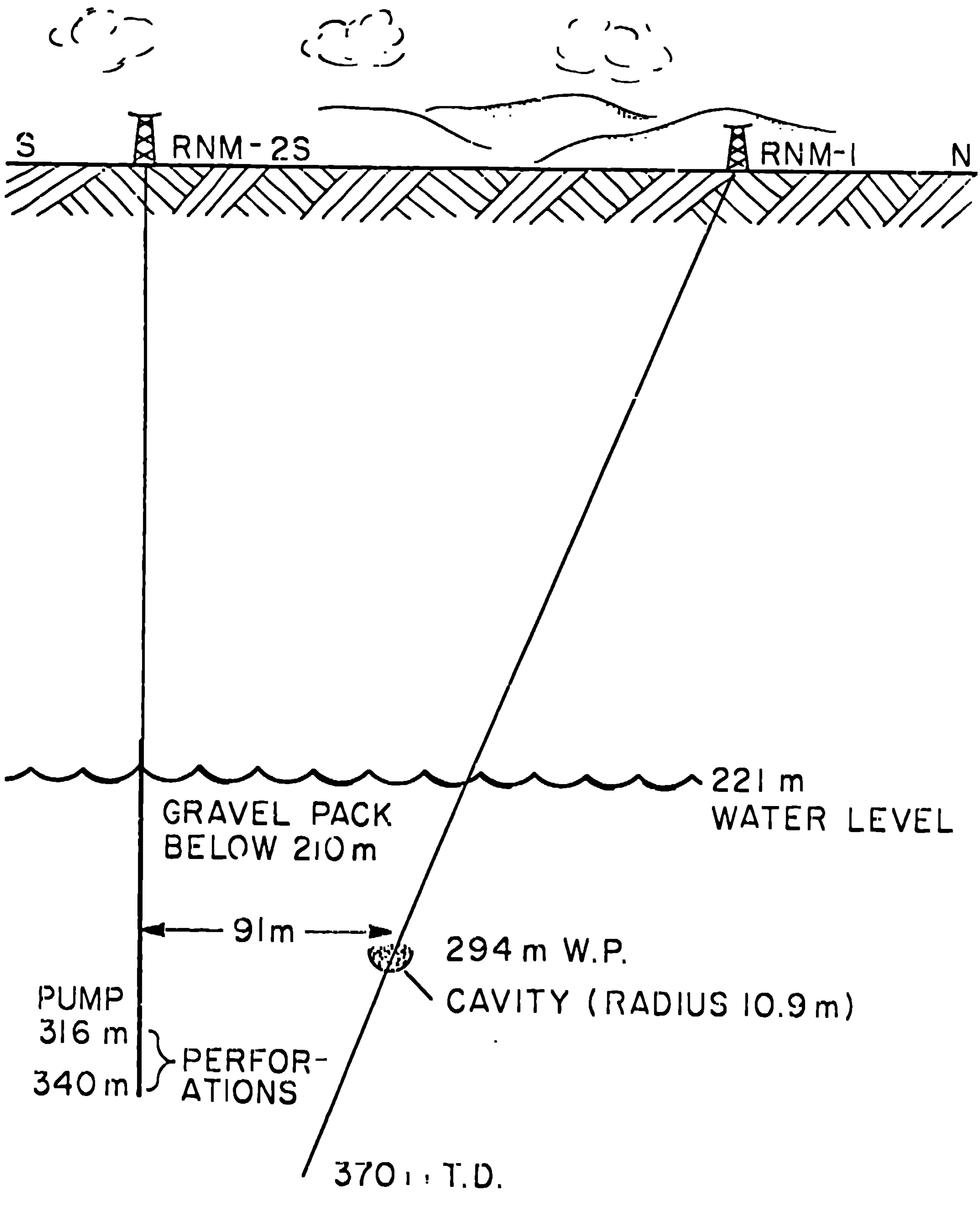

S
RNM-2S
RNM-1
N
221 m
WATER LEVEL
GRAVEL PACK
BELOW 210 m
91 m
294 m W.P.
CAVITY (RADIUS 10.9 m)
PUMP
316 m
340 m
PERFOR-
ATIONS
370 m T.D.

Figure 1

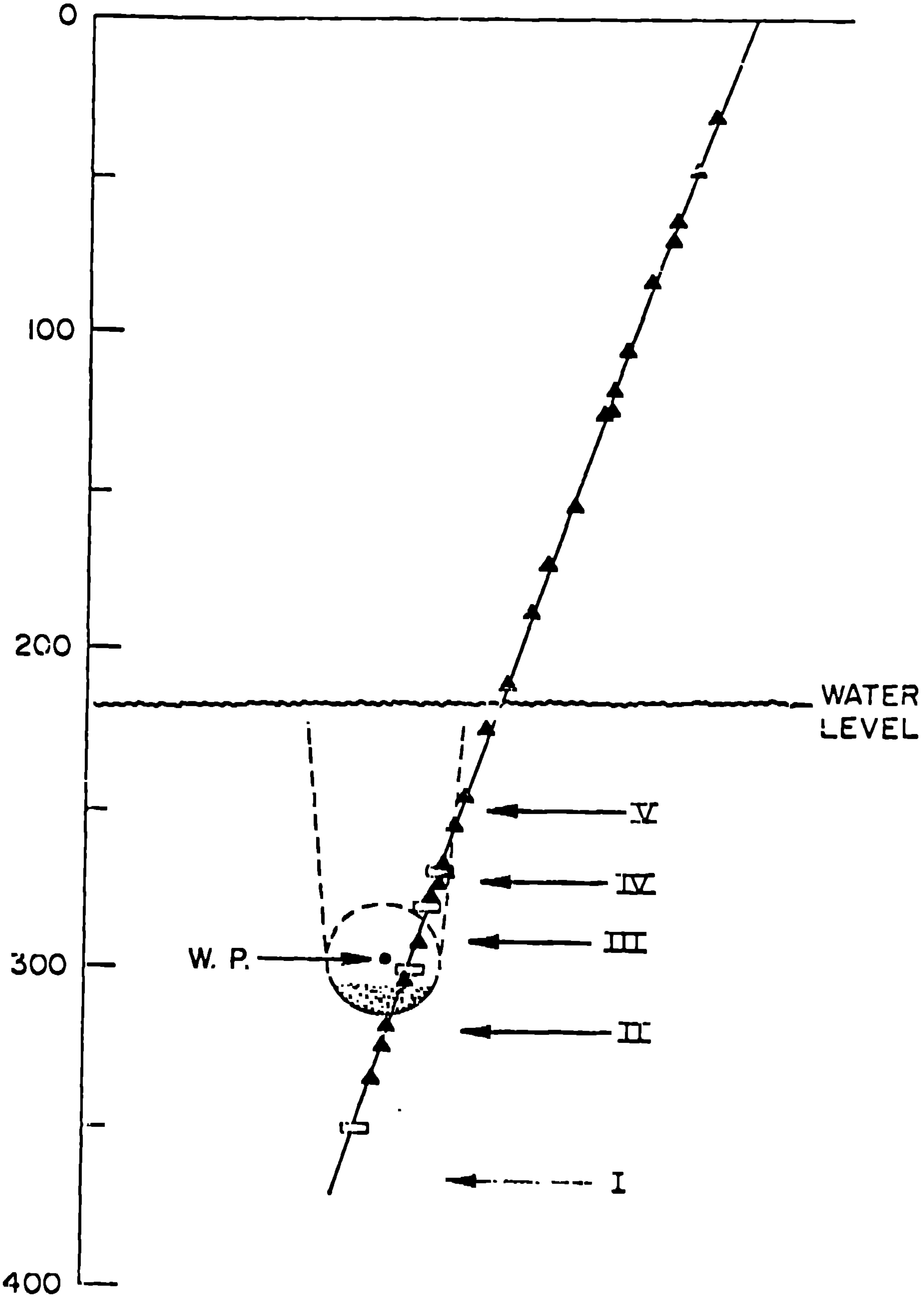


Figure 2

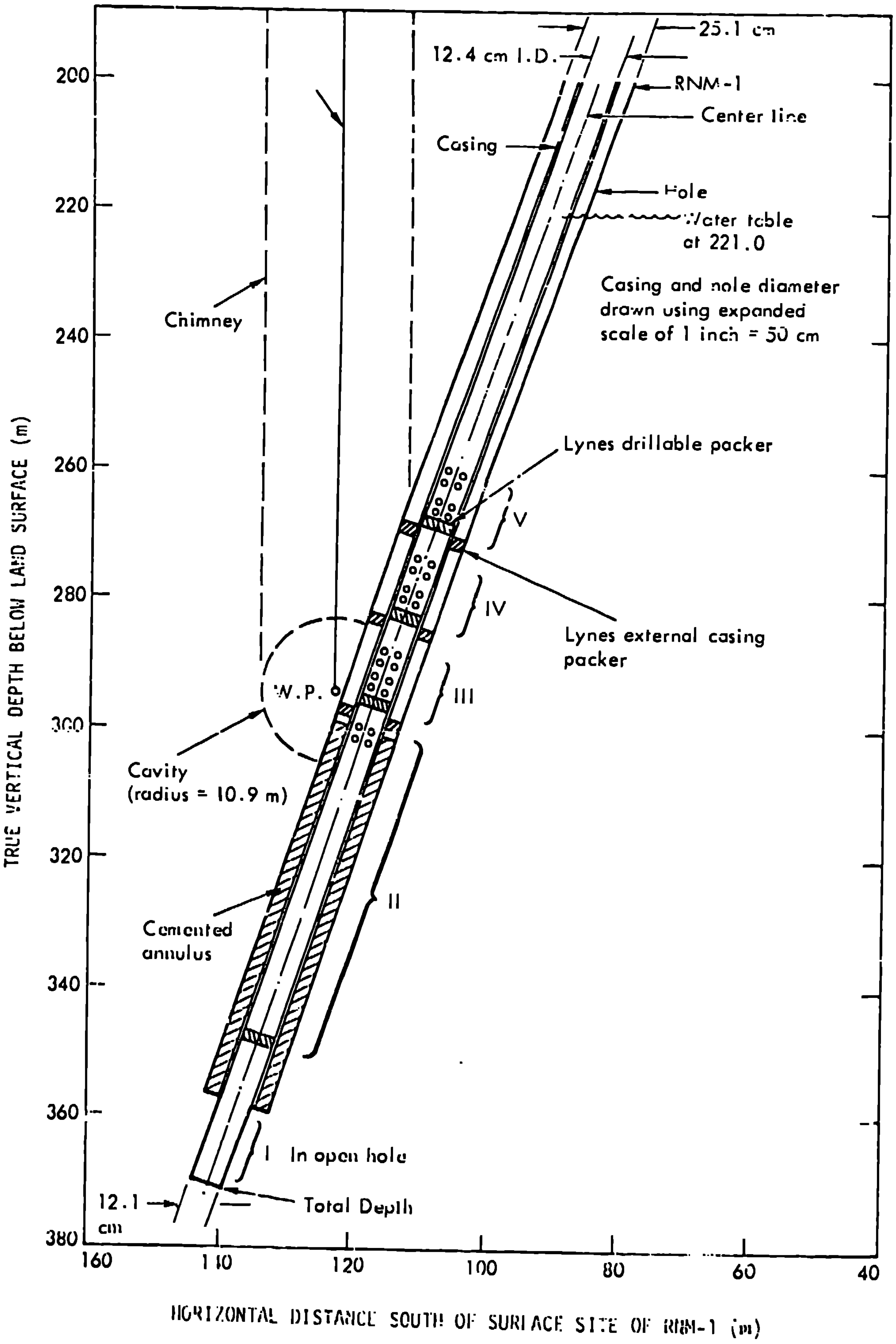
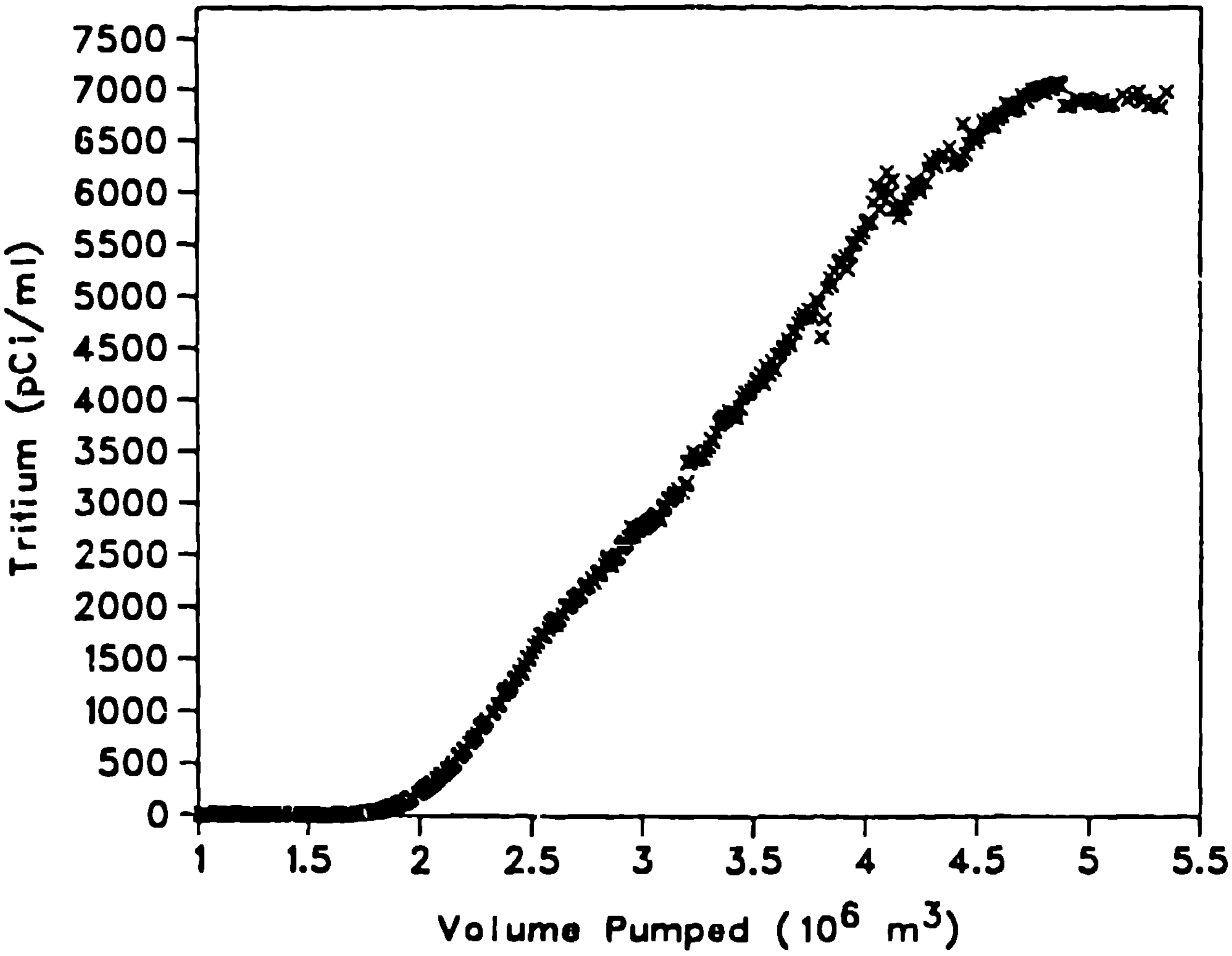
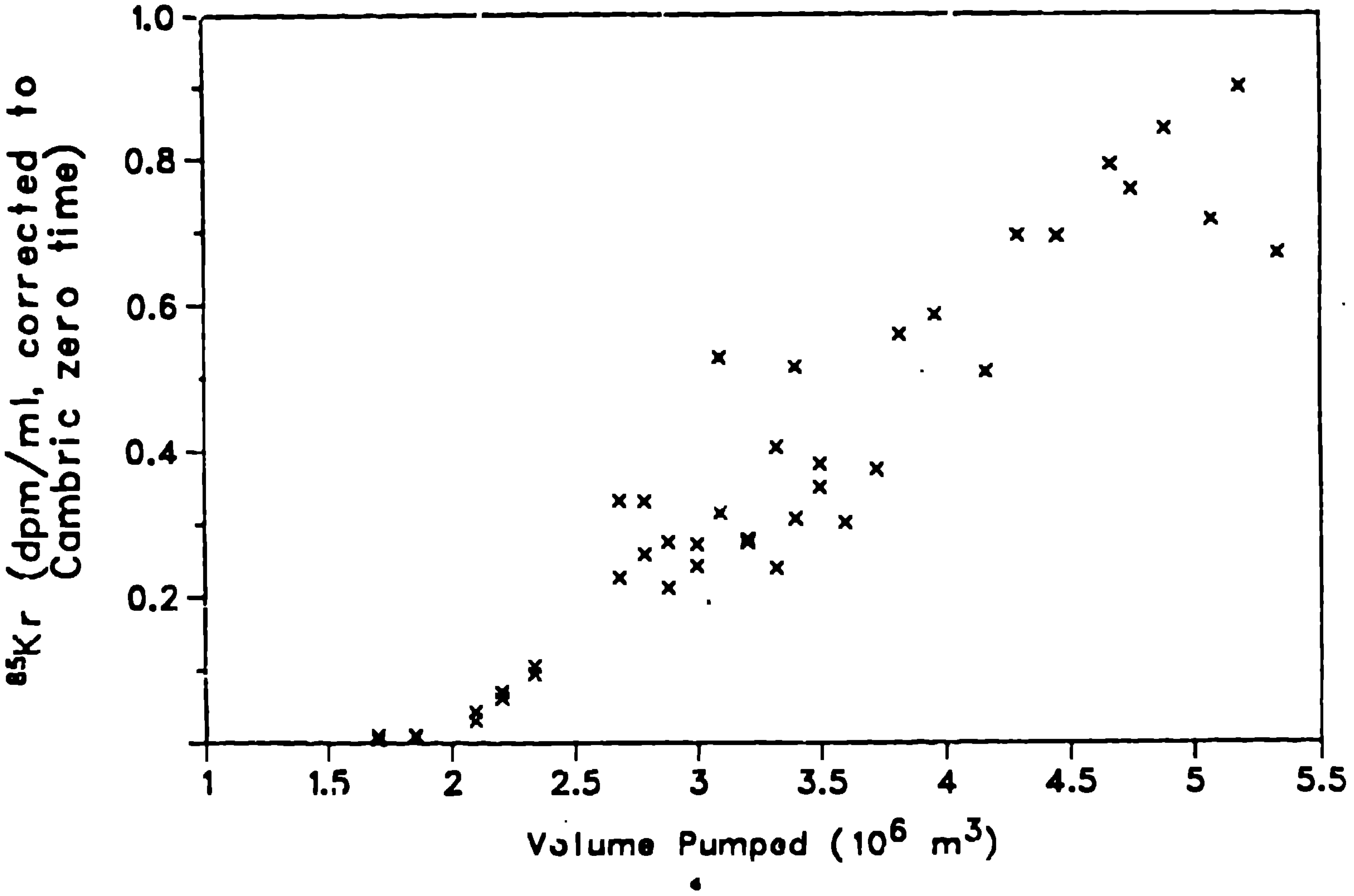

25.1 cm
12.4 cm I.D.
RNM-1
Center line
Casing
Hole
Water table at 221.0
Casing and hole diameter drawn using expanded scale of 1 inch = 50 cm
Chimney
Lynes drillable packer
V
IV
Lynes external casing packer
III
W.P.
Cavity (radius = 10.9 m)
II
Cemented annulus
I In open hole
12.1 cm
Total Depth
TRUE VERTICAL DEPTH BELOW LAND SURFACE (m)
200
220
240
260
280
300
320
340
360
380
160
140
120
100
80
60
40
HORIZONTAL DISTANCE SOUTH OF SURFACE SITE OF RNM-1 (m)

Figure [illegible]



Tritium (pCi/ml)
7500
7000
6500
6000
5500
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
1
1.5
2
2.5
3
3.5
4
4.5
5
5.5
Volume Pumped (10^6 m^3)

Figure 4



1.0
0.8
0.6
0.4
0.2
^{85}Kr (dpm/ml, corrected to Cambric zero time)
1
1.5
2
2.5
3
3.5
4
4.5
5
5.5
Volume Pumped (10^6 m^3)

Figure 5

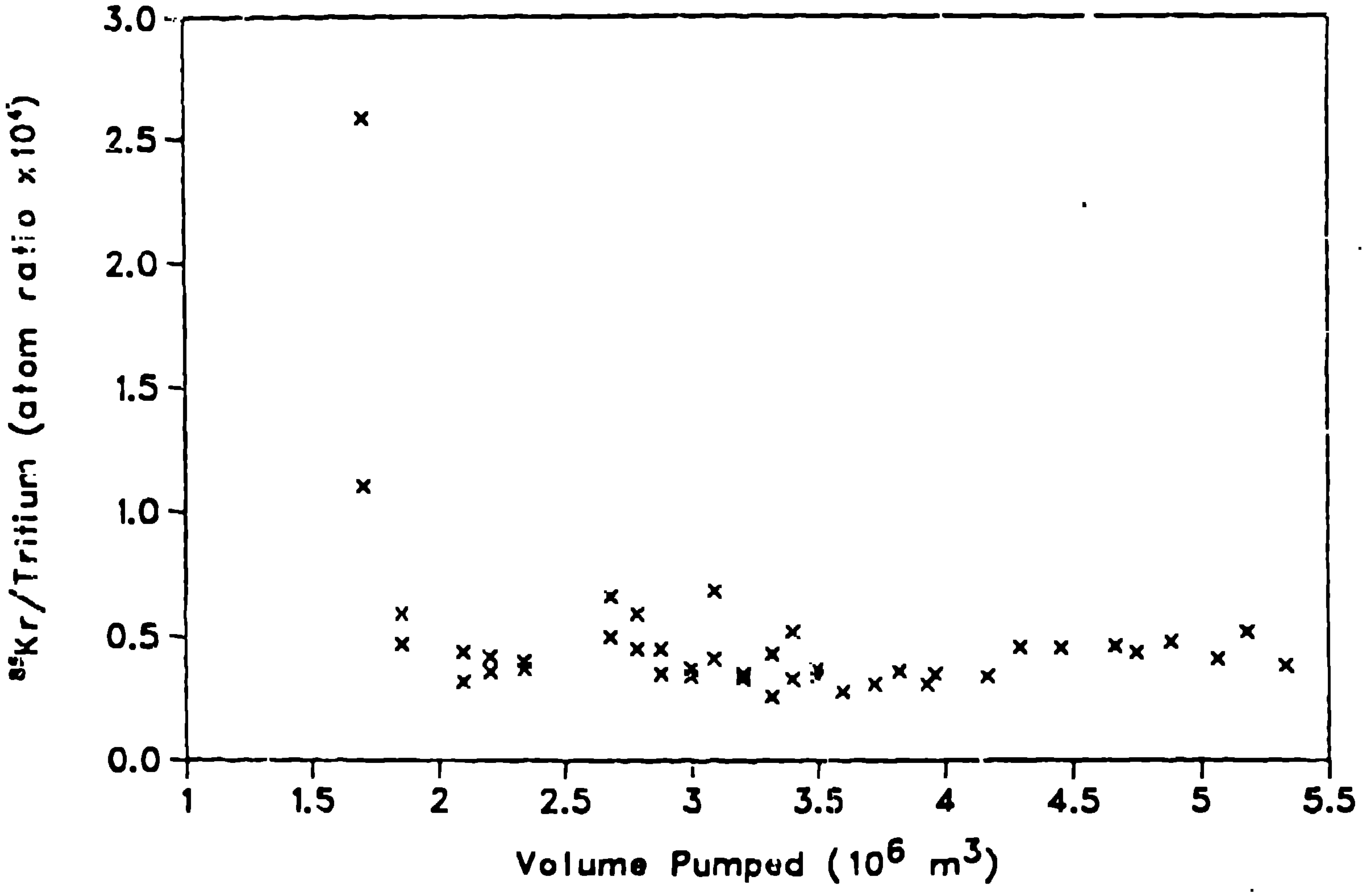

3.0
2.5
2.0
1.5
1.0
0.5
0.0
⁸⁵Kr/Tritium (atom ratio x10⁴)
1
1.5
2
2.5
3
3.5
4
4.5
5
5.5
Volume Pumped (10⁶ m³)

Figure 6

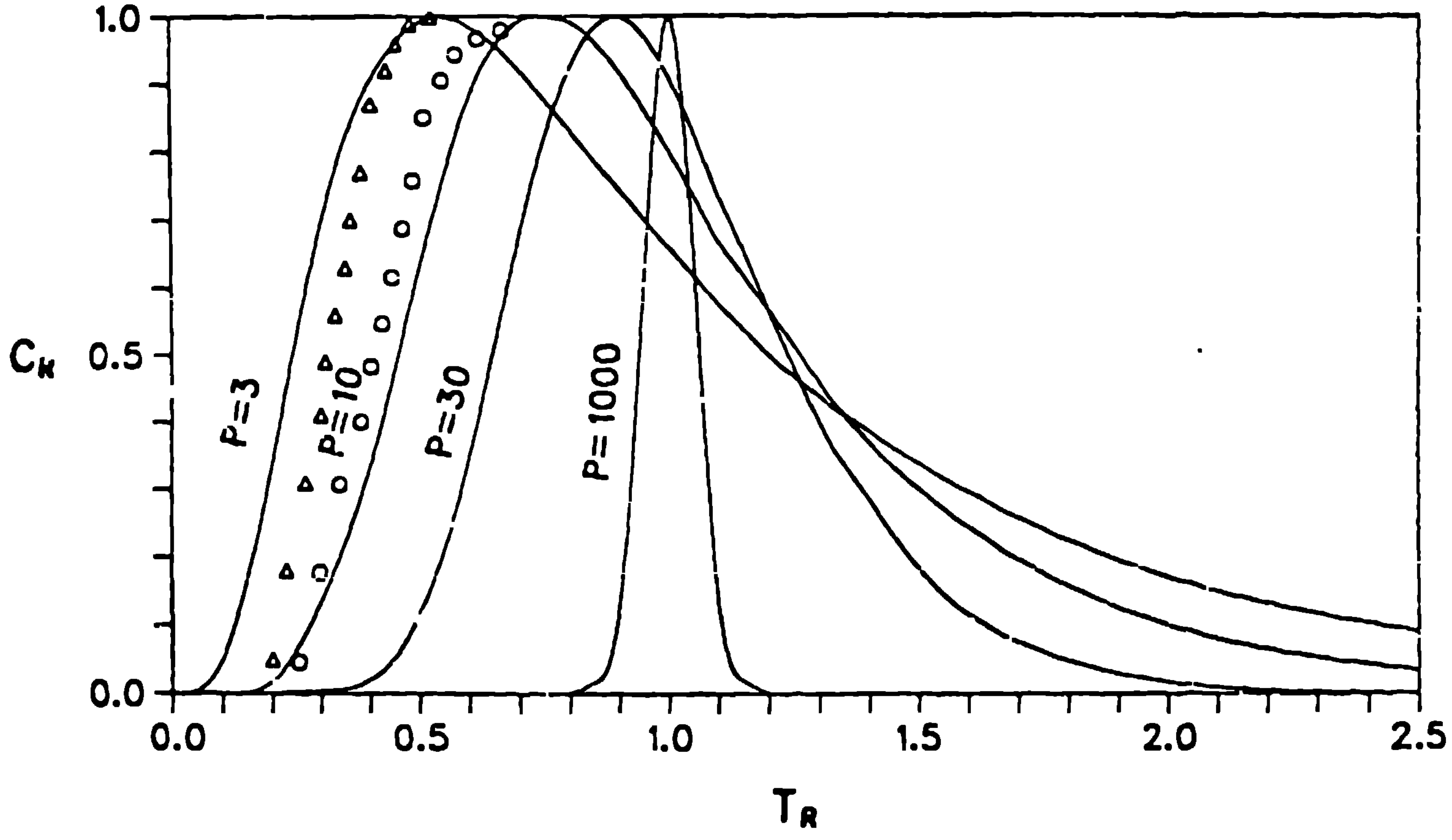


Figure 7