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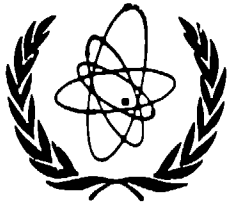


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INTERNATIONAL ATOMIC ENERGY AGENCY

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A REVIEW OF A FIELD STUDY OF RADIONUCLIDE MIGRATION FROM AN
UNDERGROUND NUCLEAR EXPLOSION AT THE NEVADA TEST SITE*

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Abstract

A REVIEW OF A FIELD STUDY OF RADIONUCLIDE MIGRATION FROM AN UNDERGROUND NUCLEAR EXPLOSION AT THE NEVADA TEST SITE. Results from a long term (9 year) field study of the distribution of radionuclides around an underground nuclear explosion cavity at the Nevada Test Site are reviewed. The goals of this Radionuclide Migration project are to examine the rates of migration underground in various media and to determine the potential for movement, both on and off the Nevada Test Site, of radioactivity from such explosions, with particular interest in possible contamination of water supplies. Initial studies were undertaken near the site of the low-yield test Cambric, which was detonated 73 m beneath the water table in tuffaceous alluvium. Solid samples were obtained from just below ground surface to 50 m below the detonation point, and water was sampled from five different regions in the vicinity of the explosion. Ten years after the test, most of the radioactivity was found to be retained in the fused debris in the cavity region and no activity above background was found 50 m below. Only tritium and ^{87}Sr were present in water in the cavity at levels greater than recommended concentration guides for water in uncontrolled areas.

A satellite well is being used to remove water 91 m from the detonation point. During seven years ($7 \times 10^6 \text{ m}^3$) of pumping, tritium, ^{86}Kr , ^{36}Cl , and ^{129}I have been detected in the water. Approximately 40% of the total tritium from the cavity region has been removed by pumping at the satellite well, and the maximum in the tritium concentration is clearly defined. Use of sensitive analytical techniques has permitted measurement of the very low concentrations of ^{36}Cl and ^{129}I present in the water. The ^{36}Cl peak precedes the tritiated water, possibly as a result of anion exclusion. Additional analyses are in progress to better define the shape of the ^{129}I concentration curve.

INTRODUCTION

A field study of the distribution of radionuclides around an underground nuclear explosion cavity was initiated in 1974, about 9 years after detonation.^{1,2} The goals of this initial experiment in the Radionuclide Migration (RNM) project were to determine the rates of migration underground in various media at the Nevada Test Site (NTS) and 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

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disposal of radioactive waste. The RNM 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 support organizations.

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

Cambric was chosen for a number of reasons, including the following: The Cambric explosion cavity is within an NTS 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, any radionuclides present in the water would constitute a potential source for transport. Sufficient tritium (^3H or T) was present to provide an easily measurable tracer for water from the cavity region. The postshot debris also contained other radionuclides whose concentrations in the rubble and groundwater from the cavity and chimney regions could be measured and compared. (A summary of the radionuclide source term at the time of re-entry is given in Table I.) The alluvium was believed to constitute a good medium for hydrologic studies because it is quite permeable and does not have large fissures or cracks through which the water might selectively flow. Finally, the

TABLE I
CAMBRIC SOURCE TERM TEN YEARS AFTER DETONATION

<u>Nuclide</u>	<u>Half Life (years)</u>	<u>Activity (Ci)</u>
^3H	12.3	3.4×10^6
^{85}Kr	10.7	4.4
^{90}Sr	29	34
^{106}Ru	1.0	2.8
^{125}Sb	2.8	3.2
^{129}I	1.59×10^7	6.6×10^{-8}
^{137}Cs	30	99
^{138}Ce	0.78	0.4
^{147}Pm	2.6	33
^{154}Eu	5.0	6.4

small yield was expected to have had little effect on the local hydrology.

The field study began with completion of a satellite well (RNM-2S) 91 m from the Cambric cavity, followed by drilling of a re-entry well (RNM-1) through the cavity itself (Fig. 1), thereby precluding the possibility of contaminating the satellite well with radioactivity from RNM-1.

Sidewall core samples were taken from RNM-1 from just below the ground surface to 50 m below the detonation point to measure the vertical distribution of radionuclides. The hole was cased and the casing perforated to permit pumping of water from different regions. In this way, variations in the radionuclide content of water from different intervals were determined, and the extent of natural radionuclide migration during the 9 years after detonation was examined.

Pumping of water from RNM-2S then induced sufficient artificial gradient to draw water from the Cambric cavity to permit study of radionuclide migration. The appearance of tritiated water (HTO) in water from this satellite well signaled the forced breakthrough of water from the Cambric cavity region. Measurement of various radionuclides present in water pumped from RNM-2S provides a means of studying radionuclide transport under field conditions.

ELEMENT DISTRIBUTION IN THE REGION OF THE CAMBRIC NUCLEAR EXPLOSION AFTER TEN YEARS

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 67 sidewall core samples was taken from RNM-1 at intervals ranging from 34 m below the surface to 37 m below the original detonation point. 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 analysis. 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 examined to determine the lithology 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).

TABLE II
EFFECTIVE DISTRIBUTION COEFFICIENTS
IN THE LOWER CAVITY REGION

Nuclide	Approximate Effective Distribution Coefficient (ml/g)
⁹⁰ Sr	10 ⁴
¹³⁷ Cs	10 ⁴
²³⁹ Pu	>10 ⁸

After sidewall core sampling had been completed and the hole had been cleaned, casing with appropriately placed, inflatable, external packers was installed.^{1, 2} The packers minimized external water movement in the annular space between the casing and the wall. Beginning at the bottom, the water in five zones (ranging from below to above the cavity) was sampled successively by isolating the zones with internal packers and perforating the casing. 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 above background 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 ⁸⁵Kr and tritium were found in the collapsed 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 ⁸⁵Kr 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 ⁹⁰Sr at levels higher than the concentration guides^{3, 4} (CG) for water in uncontrolled areas.

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, E_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 are retained below the water table and that the fraction of a nuclide not in the water was in the solid.)

TABLE III
 REPRESENTATIVE ACTIVITY LEVELS IN CAMBRIC
 WATER SAMPLES TEN YEARS AFTER DETONATION

Zone	Tritium Activity Level ($\mu\text{Ci}/\text{ml}$)	Activity Level (dpm/ml)					
		^{85}Kr	^{90}Sr	^{106}Ru	^{125}Sb	^{137}Cs	^{239}Pu
I (below cavity)	bg ^a	bg	bg	bg	bg	bg	bg
II (lower cavity)	6.1	800	8	11	5	1.6	≤ 0.003
III (upper cavity)	3.8	1200	5	4	2	1.4	≤ 0.003
IV (chimney)	0.084	70	5	nd ^b	nd	0.8	nd
V (adjacent to chimney)	0.028	20	0.2	nd	nd	0.2	nd
CG ^c	2×10^{-4}	---	0.018	22.2	222	44.4	11.1

^abg = no activity detectable above background levels.

^bnd = not detected.

^cCG = recommended^{1,2} concentration guide applicable to water in uncontrolled areas. Values for tritium and ^{90}Sr are public drinking water standards established in Ref. 4.

$$E_d = \frac{(A_X/A_T)_{\text{Cambric}} - (A_X/A_T)_{\text{water}}}{(A_X/A_T)_{\text{water}}} = \frac{(A_X)_{\text{Cambric}}(A_T)_{\text{water}}}{(A_X)_{\text{water}}(A_T)_{\text{Cambric}}} - 1$$

Substituting $\frac{A_{T \text{ water}}}{A_{T \text{ Cambric}}} \geq 0.999 \approx 1$ (Ref. 2)

$$E_d = \frac{A_X \text{ Cambric}}{A_X \text{ water}} - 1$$

Then

$$E_d = \frac{A_X \text{ solid}}{A_X \text{ water}} \approx K_d \frac{\mu\text{g solid}}{\text{cc water}}$$

or

$$K_d \approx E_d \frac{\text{wt.}\% \text{ water}}{100 - \text{wt.}\% \text{ water}}$$

where K_d is the distribution coefficient.

The nuclides ^{90}Sr , ^{106}Ru , ^{125}Sb , ^{137}Cs , ^{239}Pu , 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).

Additional information describing early work at RNM-1 is given in Refs. 1 and 2.

TRANSPORT OF RADIONUCLIDES TO THE SATELLITE WELL

The satellite well RNM-2S is located 91 m from 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 detected after a total of 1.44 x 10⁶ m³ of water had been pumped from RNM-2S.

The tritium concentration in water from RNM-2S reached a maximum just below the maximum permissible³ concentration (3nCi/ml) for drinking water in controlled areas and is now decreasing (Fig. 2). Pumping of 7.09 x 10⁶ m³ of water from RNM-2S (by early October 1982) removed 42% of the tritium available from Cambric. Figure 3 shows a comparison of the transport of tritiated water to the satellite well with the calculations of Sauty⁵ for instantaneous tracer injection in a radial, converging flow field, similar to the RNM experiment. The dimensionless time is given on the upper abscissa. A dimensionless, time T_r , of one corresponds to the time required to pump the volume of water contained in a cylinder whose radius is the distance from the satellite well to the source. The dimensionless concentration, C_r , is the concentration relative to the maximum concentration defined by the data. This treatment invokes symmetry and treats the source as an annular ring.

The shape of the elution curve depends on the Peclet number, which is inversely proportional to the dispersivity. An excellent fit to the data was obtained with a Peclet number of 10, corresponding to a dispersivity of 9.1 m. This result can be compared with the data presented by Borg et al.,⁶ who reported longitudinal dispersivity values that were estimated by calibrating mathematical models for transport against observed transport in various field studies. Values ranged from 11.6 to 91 m for a wide variety of lithologies; the value was 21.3 m for a sand or gravel deposit -- the lithology that probably most closely resembles the tuffaceous alluvium of the current experiment.

Chlorine-36 produced by the Cambric event has been detected in water samples from RNM-2S using accelerator-based mass spectrometry. The technique is described in Ref. 7. A comparison of the concentrations of tritiated water and ³⁶Cl is given in Fig. 4. As for tritium, the two-dimensional solutions of Sauty⁵ were used to fit the ³⁶Cl data and an excellent fit was obtained with Peclet number 10 (Fig. 5).

TABLE IV
RETENTION FACTORS FROM CAMBRIC WATER SAMPLES

Zone	⁹⁰ Sr	¹⁰⁶ Ru	¹²⁵ Sb	¹³⁷ Cs	¹⁴⁷ Pm	²³⁹ Pu
I (below cavi)	bg ^a	bg	bg	bg	bg	bg
II (lower cavity)	2.1x10 ¹	1.0x10 ¹	2.9x10 ¹	2.5x10 ⁰	≥10 ⁰	≥3.2x10 ¹
III (upper cavity)	1.6x10 ¹	1.9x10 ¹	3.6x10 ¹	1.8x10 ⁰	nd	≥1.9 x 10 ¹
IV (chimney)	3.9x10 ¹	nd ^b	nd	6.6x10 ¹	nd	nd
V (adjacent to chimney)	3.1x10 ¹	nd	nd	1.1x10 ¹	nd	nd

^abg = no activity detectable above background levels.

^bnd = not detected.

The breakthrough of ³⁶Cl appears to occur earlier than that of tritium, and the maximum in the chloride concentration clearly occurs before that of tritium. The phenomenon whereby anions are eluted earlier than cations or neutral species from packed columns has been observed in soil chemistry studies^{8,9} and column chromatography and is called anion exclusion. The effect has been shown to occur in soils containing smectites, and it correlates fairly well with the cation exchange capacity of the soil. Anion exclusion is due to the negative surface charge on aluminosilicates which repels anions, thereby effectively constricting the pores and making the intragranular porosity of the mineral unavailable to negatively charged species.

A method has been devised for determining the concentration of ¹²⁹I in RNM waters by neutron activation and counting of the ¹³⁰I product.¹⁰ Although the concentrations are well below the concentration guide³ for water in an uncontrolled area (the concentration of ¹²⁹I in RNM-1 water was 1.1 x 10⁻⁶ μCi/ml; the CG is 6 x 10⁻⁶ μCi/ml), the observation is significant because ¹²⁹I is considered a hazardous radionuclide in the storage of nuclear waste.¹¹ The high ranking of ¹²⁹I is due to its biological activity and lack of sorption on most minerals.

The concentration of ¹²⁹I was determined in samples taken from RNM-1 before pumping started on RNM-2S. The concentration of ¹²⁹I in water removed from the lower cavity region is ~2.6 x 10¹⁴ atoms/liter. The highest concentration of ¹²⁹I measured in the RNM-2S samples was 7.6 x 10¹⁰ atoms/liter. The iodine and tritium concentrations for RNM-2S water are plotted in Fig. 6. The breakthroughs in the two curves occur at approximately the same volume; however, it is not clear that the maxima coincide. If the iodine were present as an anionic species, the breakthrough and peak should precede the tritium as they do for ³⁶Cl.

A stability diagram for iodine (Fig. 7) was calculated using the half-cell potentials of Kahn and Kleinberg¹² for 10^{-7} M iodine. The complex behavior of iodine in dilute solutions was discussed by Kahn and Kleinberg. The dashed line in Fig. 7 represents the Eh of fully oxygenated water. The pH of the groundwater around Cambria is about 7.5 to 8. Bearing in mind that the concentration of iodine in natural water is approximately 10^{-7} M, if the groundwater were air-saturated, the dominant species should be HIO, a neutral species. This would be a possible explanation for a smaller anion exclusion effect.

Relative to the concentrations measured in RNM-1, only about one quarter as much ^{129}I as tritium has been recovered at RNM-2S. Whether this is due to retardation, peak broadening resulting from multiple species, loss of iodine to organic material, or some other mechanism of removal remains to be seen. The loss of iodine tracer in elutions through soil columns has been observed in laboratory experiments.¹³ Recoveries of ^{131}I from 44 to 15% were obtained in these experiments for three soils, and increasing quantities of clay, silt, and organic matter appeared to reduce the recovery of iodine. This loss of iodine may be related to the presence of HIO which is believed¹² to disproportionate to form a reactive unidentified species which may react with organic material or perhaps even silica.

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. Analyses of these samples show the presence of ^{85}Kr (Fig. 8), which was transported from RNM-1 to RNM-2S in the water. The $^{85}\text{Kr}/\text{T}$ atom ratios are relatively constant at a value of approximately 0.4×10^{-4} , considerably lower than that of 1.22×10^{-4} calculated for Cambria. So far the reason for this is not known; however, it may 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 (chimney region) and V (just outside the chimney); 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 1000, presumably as a result of pumping at RNM-2S; however, the ^{85}Kr concentration is apparently decreasing more slowly than the tritium concentration. The $^{85}\text{Kr}/\text{T}$ ratios obtained for samples collected from RNM-1 after pumping began at RNM-2S are consistently greater than the calculated ratio for Cambria. Other data show that the concentrations of ^{90}Sr and ^{137}Cs in water pumped from RNM-1 have also decreased significantly (factors of 100 and 15, respectively) as a result of pumping at RNM-2S.

Twenty-four 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 measured. With the possible exception of a very small amount of ^{106}Ru (<1% of that produced in Cambric),¹⁴ no gamma-emitting nuclides have been identified in these samples. Radiochemical analyses of water samples for ^{137}Cs and ^{90}Sr have also given negative results.

SUMMARY AND CONCLUSIONS

* The available source term for radionuclides in the region of the Cambric nuclear explosion was determined. Drillback cores were obtained and analyzed, and water was pumped from several vertical zones and analyzed. Most of the radioactivity produced in the detonation 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 water in uncontrolled areas.

During seven years ($7 \times 10^4 \text{ m}^3$ of water) of pumping from a satellite well located 91 m from the Cambric cavity, tritium, which is present as HTO and chemically the same as the water, ^{85}Kr , which seems to be dissolved in the water, ^{36}Cl and ^{129}I , presumably as anionic or possibly neutral species, have been observed in water removed from this well. There is some evidence for the possible transport of minute amounts of 1.0-year ^{106}Ru (< 1% of that produced in the explosion). The results from RNM-2S indicate that, in general, radionuclide sorption is sufficiently high to preclude the transport of most nuclides, particularly cationic species, from the original cavity to the satellite well in the near future. Pumping and radioassay of water from the satellite well will be continued in order to investigate the possible arrival of other poorly sorbing species.

ACKNOWLEDGMENTS

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TABLE V
COMPARISON OF ^{85}Kr AND TRITIUM
CONCENTRATIONS IN RNM-1 WATER

Entry	Date	Volume Water ^a (10^6 m ³)	Concentration ^b		$^{85}\text{Kr}/\text{T}$ Atom Ratio	
			T (10^6 atoms/ml)	^{85}Kr (10^6 atoms/ml)	($\times 10^6$)	R ^c
Original - Zone IV	08/08/75	0	3100	570	1.8	1.5
Original - Zone V	08/14/75	0	790	110	1.4	1.2
Zones IV + V	10/04/77	1.17	66	610	92	75
Zones IV + V	11/30/77	1.34	41	49	12	10
Zones IV + V	09/04/79	3.50	5.4	4.6	8.5	7
Zones IV + V	10/05/81	5.89	2.7	1.5	5.6	5
Zones IV + V	04/01/82	5.48	1.7	0.55	3.3	3

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

^bValues corrected to Cambria zero time.

^cMeasured $^{85}\text{Kr}/\text{T}$ divided by $^{85}\text{Kr}/\text{T}$ calculated for Cambria (1.22×10^{-6}).

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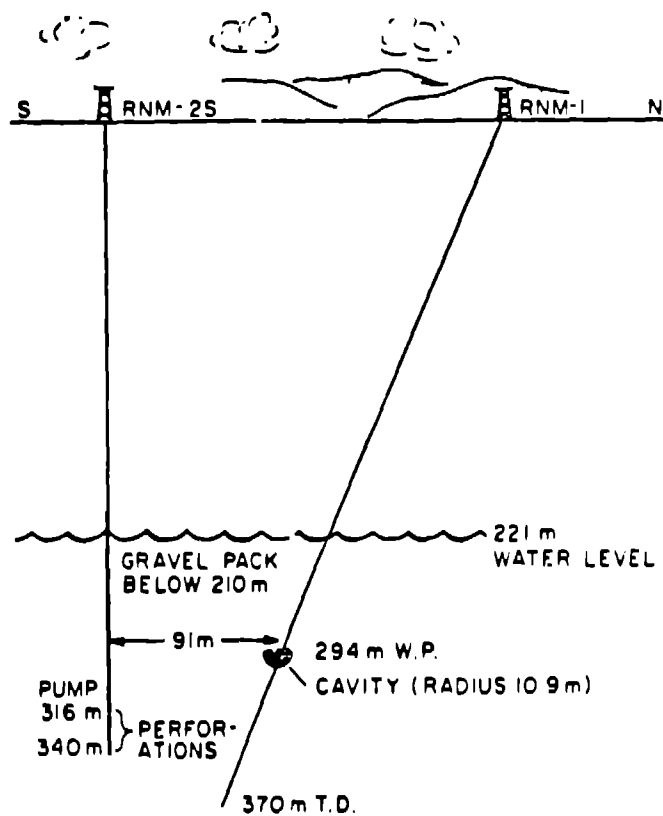


FIG. 1. Schematic of the Cambric site.

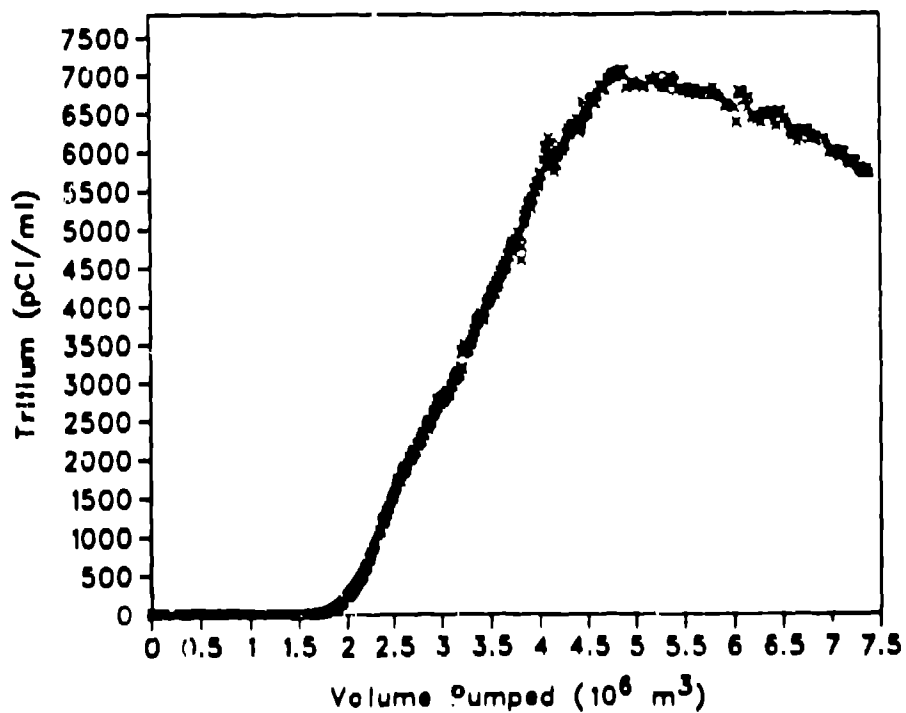


FIG. 2. Tritium concentration in water from RNM-2S, corrected to Cambric zero time. Irregularities correspond to periods during which the pump was not operating.

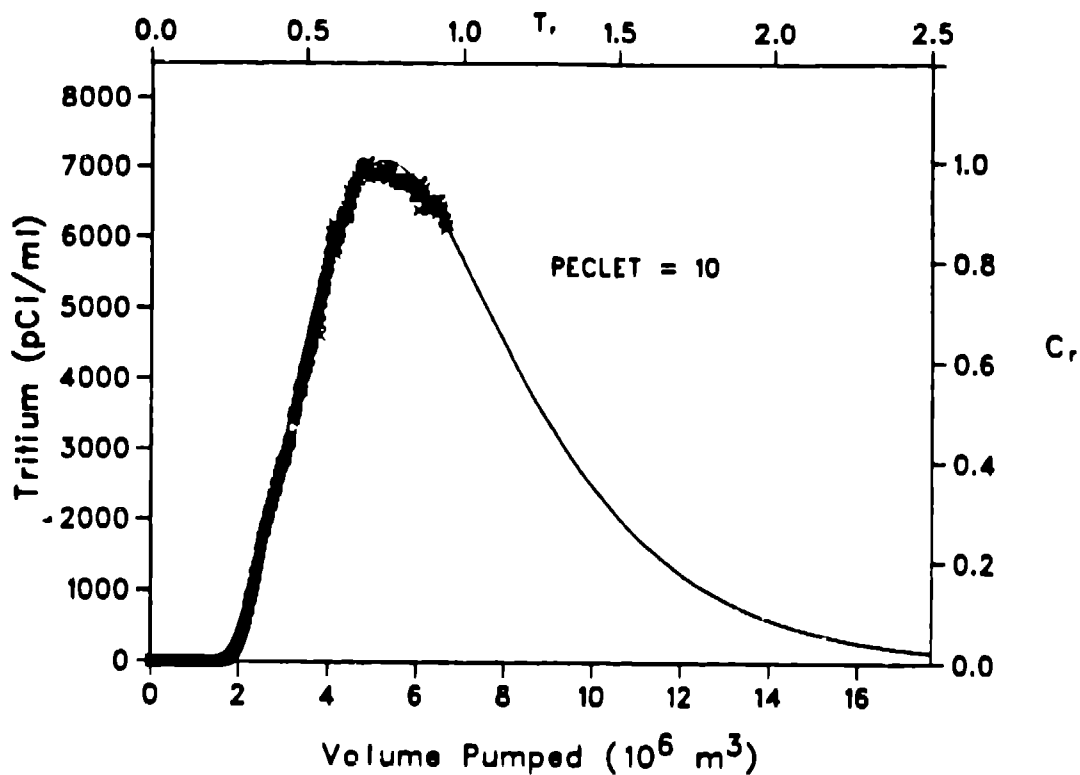


FIG. 3. Tritium concentration data for RNM-2S water and calculated elution of tracer after instantaneous tracer injection in a radially converging flow field for Peclet number 10.

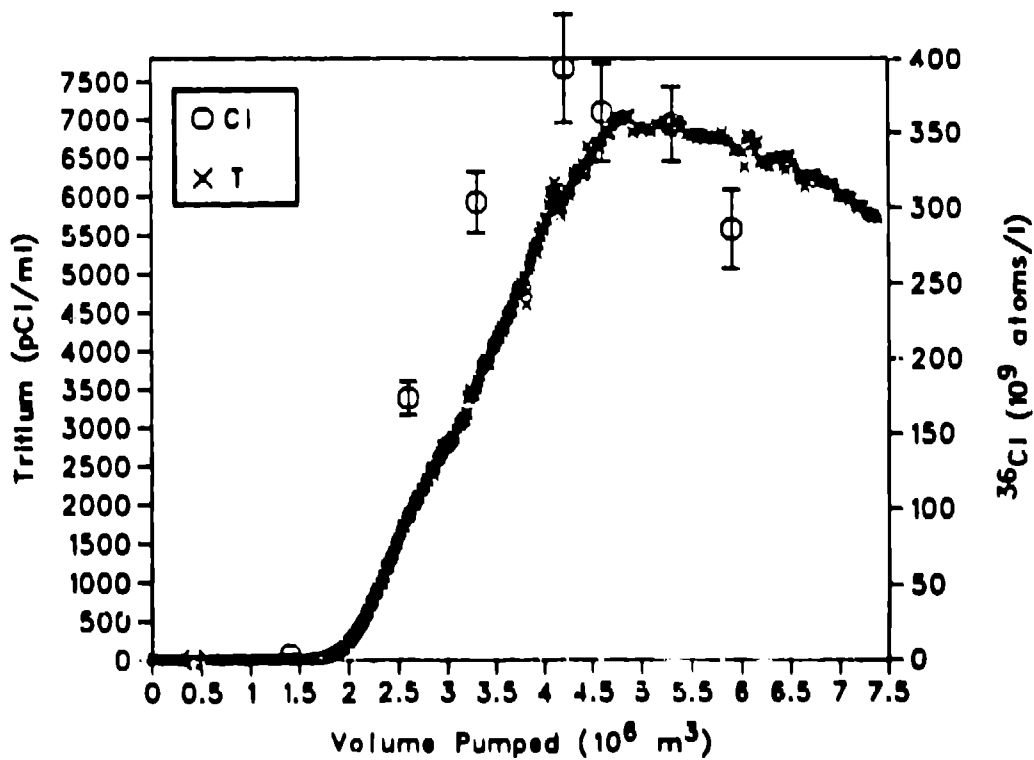


FIG. 4. Concentrations of ^{36}Cl and tritium in RNM-2S. The error bars are the standard deviations for single measurements; they were obtained from the errors associated with the measurements and estimated uncertainties for various parameters entering into the calculation.

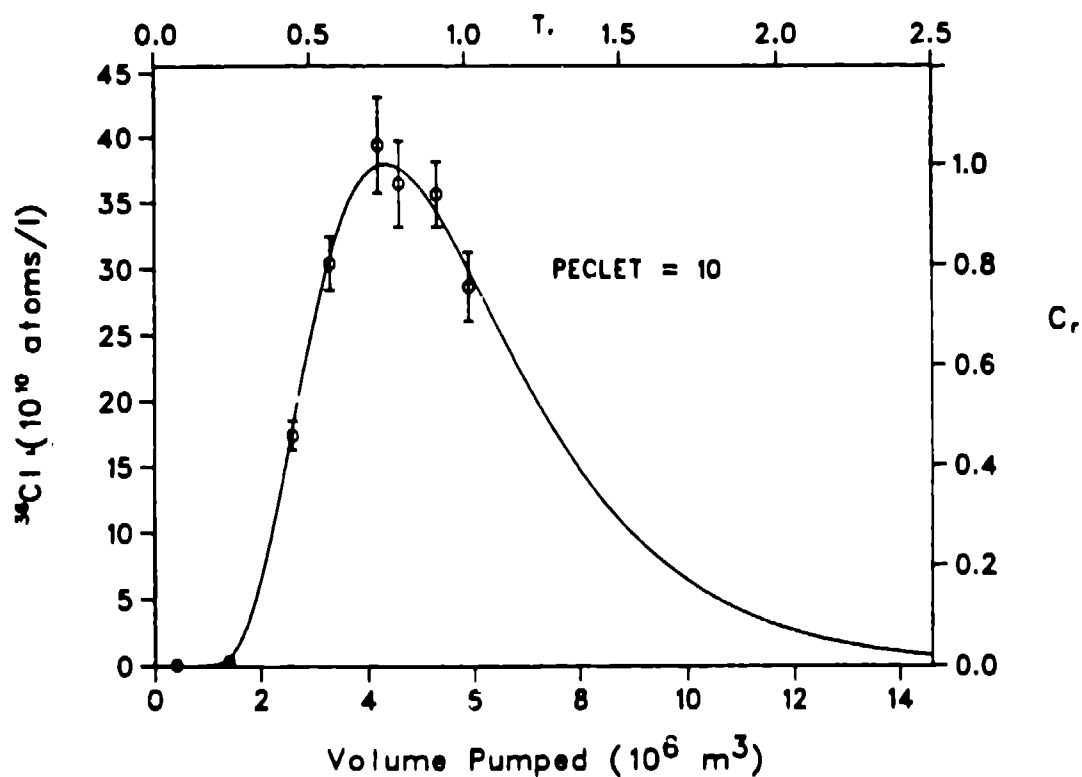


FIG. 5. Chlorine-36 concentration data for RNM-2S water and calculated elution of tracer after instantaneous tracer injection in a radially converging flow field for Peclet number 10.

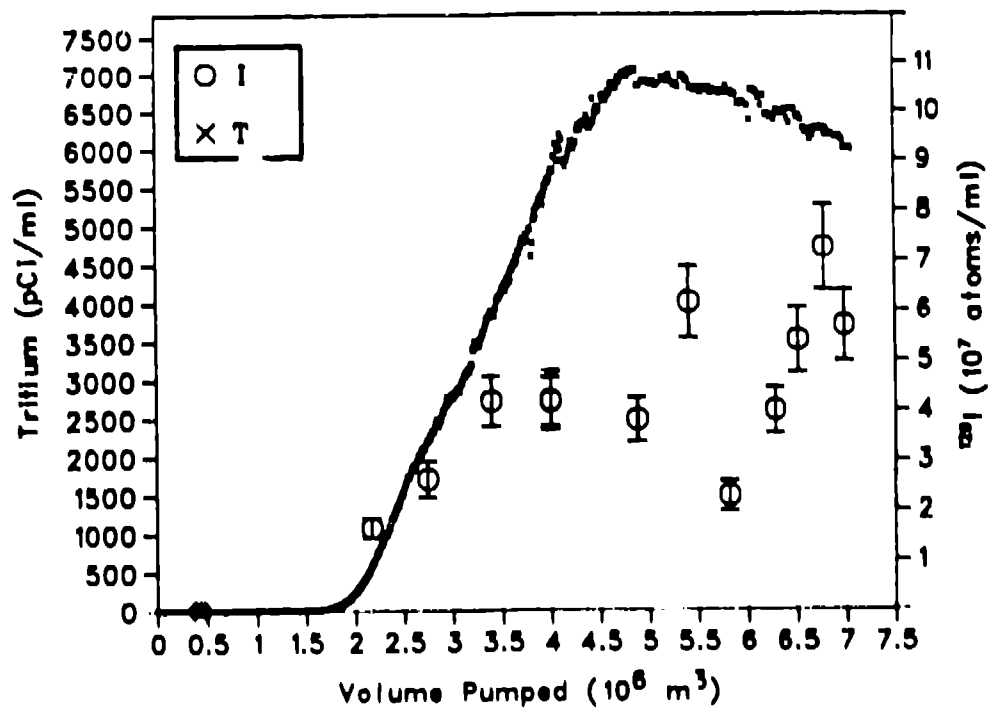


FIG. 6. Concentrations of ^{131}I and tritium in RNM-2S water.

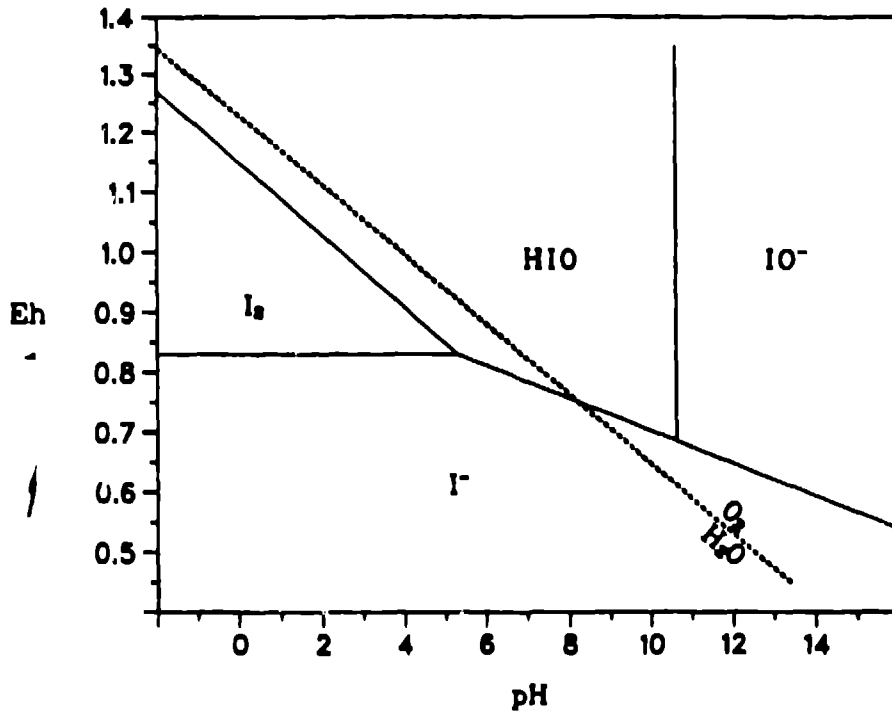


FIG. 7. Iodine stability diagram (total iodine concentration 10^{-7} M).

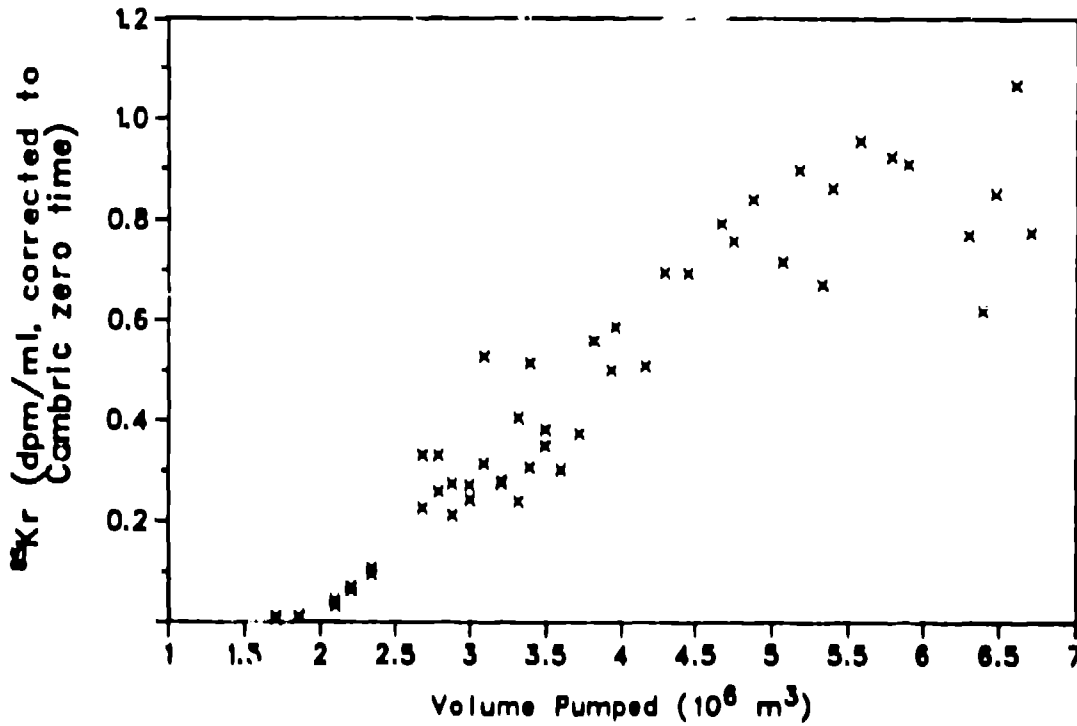


FIG. 8. Krypton-85 concentration in water from RNM-2S, corrected to Cambria zero time.