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MIGRATION IN ALLUVIUM OF CHLORINE-36 AND TRITIUM FROM AN UNDERGROUND NUCLEAR TEST[1]

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SUMMARY: This article describes a field experiment studying the migration in alluvium of radioactive elements away from an underground nuclear explosion at the Nevada Test Site in the United States. Nuclides detected in the pumped water are tritium, chlorine-36, iodine-129, and krypton-85 all at levels below the maximum permissible concentration for drinking water in controlled areas. The chlorine-36 elution curve precedes that of tritium and is due to an anion exclusion process. A conventional two-dimensional convection-diffusion equation does not fully describe the elution curves for tritium and chlorine-36; the tailing of the curves is longer than predicted. Successful modeling of this experiment will be important for validating codes and models to be used in the high-level nuclear waste program.

KEY WORDS: Radionuclide Migration, Chlorine-36, Tritium, Field Experiment.

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INTRODUCTION:

In 1974, the Radionuclide Migration (RNM) project was started at the Nevada Test Site (NTS) to determine the potential for movement of radioactivity away from underground nuclear explosions.

The first field experiment in this project is a long-term single-well pumping test in which the cavity of a nuclear explosion is treated as the slug-injection point and the explosion products are the tracers. The site of the 0.75-kt nuclear explosion called Cambric was chosen for this field experiment. This experiment should be close to ideal for modeling because 1) the small yield of the explosion should not have influenced the natural hydrology of the area to any great extent, 2) sufficient tritium and other radionuclides are present to serve as tracers for both the water flow and the sorbing nuclides, and 3) alluvium is perhaps the simplest and most uniform medium at the NTS for such an experiment and therefore should simplify the modeling process.

EXPERIMENTAL:

Two wells, as shown in Figure 1, were drilled 91 m apart for this test. RNM-2S was drilled first so that it would not be contaminated by the drilling of RNM-1, which was drilled through the explosion cavity. The casing in RNM-2S was perforated at depths of 316 to 340 m, and a pump was installed above these perforations. RNM-2S was pumped from October 1975 to October 1977 at the rate of 1 m^3/min; since that time it has been pumped at a rate of 2.3 m^3/min. A pump installed above the packers and perforations in RNM-1 allows water to be pumped from the area immediately above the cavity. RNM-1 has been pumped yearly to obtain information on the changes of the radionuclide concentrations in the source. Additional details on the construction of the wells can be found in Hoffman, Stone and Dudley (1) and Hoffman et al. (2).

The alluvium in this area of the NTS extends downward for approximately 700 m before volcanic rocks are encountered. These in turn extend an additional 600 m to the Paleozoic rocks. Characterization of the alluvium at the Cambric site was done by studying

13 sidewall samples taken between 103.6 and 319.9 m during the drilling of well RNM-1. Sampled locations are shown in Figure 1. Information was also obtained from cutting samples taken during the drilling of RNM-2S and those from an exploratory hole 500 m distant. The samples from these three holes are quite uniform and are composed of tuffaceous sand and gravel (1,3). Lithologies of the sand particles indicate that they were derived mainly from Tertiary volcanic rocks. Only a small portion of the clasts originated from Paleozoic rocks . One sidewall sample, however, was different. The sample from 292-m vertical depth in RNM-1 was composed of silty, bluish-gray clay.

In the early part of the pumping, samples were taken weekly for tritium analysis, but in July 1984, the sampling became monthly. The samples are analyzed both by Reynolds Electric Company at the NTS and by us at Los Alamos. Liquid scintillation counting is used by both laboratories and good agreement is obtained (1,4-9). Water samples are taken in pre-evacuated stainless steel tubes that are connected directly to the main water line from well RNM-2S. In this way the content of the water is not influenced by the atmosphere. The water in these samples is analyzed for krypton-85 and tritiated water. Plastic-lined barrels holding 208 l are also filled once a month, but no effort is made to prevent this water from being exposed to the air as is done with the stainless steel tubes. These large samples are for future analysis. In some cases they are boiled down to dryness and the residue is examined with Ge(Li) detectors for gamma-emitting radioisotopes. Portions of some of the large samples are also used for chlorine-36 and iodine-129 analyses. The ratios of chlorine-36 to chlorine were determined by accelerator mass spectrometry at the University of Rochester. A complete description of the analytical method can be found in Elmore et al. (10). Chlorine-36 contents were then calculated from the known amount of chlorine added to each sample. The results of tritium and chlorine-36 analyses are discussed in this article.

RESULTS:

Tritium, as tritiated water, is by far the most abundant of the radioactive materials in the water being pumped from well RNM-2S. It is inexpensive to analyze and is generally considered to be an ideal groundwater tracer because it travels as the water molecules. The tritium results and discussion will be developed before those of chlorine-36 so that there will be a baseline against which the chlorine-36 results can be compared.

The individual values of the tritium concentrations are far too numerous to list here; the reader is directed to the Los Alamos reports in references 4-9. All the tritium results are included in Figure 2, which shows the tritium elution curve for a period of more than 11 years. Discontinuities in the curve occur during short periods when the pump was not operating. By September 1986, 75% of the original tritium present at Cambric after the detonation had been pumped out through RNM-2S.

The chlorine-36 content for some of these same waters are listed in Table 1. Because of the high cost for analysis of low-level chlorine-36 samples, their number is much smaller than the number of tritium analyses.

The chlorine-36 concentrations are also included in Figure 2 so that they can be compared to the tritium data. The ordinates for both tritium and chlorine-36 were drawn so that each set of data was best fit by the same numerical solution, as shown later in Figures 4 and 5.

DISCUSSION:

A comparison of the elution data for chlorine-36 with that of tritium in Figure 2 indicates that the breakthrough of the chlorine-36 appears first and the maximum in the chlorine-36 data also occurs before that of tritium. This phenomenon of anions such as chloride being eluted before cations or neutral species such as tritiated water has been observed in soil chemistry studies by Thomas and Swoboda (11) and has been given the descriptive name of anion exclusion. Anions, being of the same charge as any clays or zeolites in the soil, are repelled by clays and perhaps zeolites and are effectively

TABLE 1

CHLORINE-36 CONCENTRATIONS IN CAMBRIC WATERS

Volume Pumped (10^6 m^3)	^{36}Cl (10^{11}atoms/l)	Volume Pumped (10^6 m^3)	^{36}Cl (10^{11}atoms/l)
0.41	0.01 ± 0.002	4.6	3.61 ± 0.33
1.4	0.03 ± 0.006	4.88	3.87 ± 0.29
2.6	1.74 ± 0.11	5.3	3.56 ± 0.25
2.61	2.03 ± 0.17	5.58	2.96 ± 0.26
2.99	2.89 ± 0.16	5.9	2.86 ± 0.26
3.3	3.04 ± 0.20	6.59	2.73 ± 0.24
3.39	4.04 ± 0.27	7.77	2.22 ± 0.20
3.81	3.95 ± 0.26	8.94	1.64 ± 0.17
4.17	5.12 ± 0.39	10.19	1.18 ± 0.10
4.2	3.94 ± 0.37	10.87	1.10 ± 0.06
4.6	3.66 ± 0.26	12.0	1.01 ± 0.05

prevented from entering into the intergranular porosity of the soil particles.

To calculate the value of anion exclusion in our ongoing experiment, it is necessary to fit separately the elution data for chlorine-36 and tritium to a numerical equation such as the two-dimensional solutions of Sauty (12) for instantaneous tracer injection in a radial converging flow field. The difference in the two generated curves is then used to calculate an exclusion volume.

The early part of the chlorine-36 and tritium elution curves through the maxima in Figure 2 have been modeled by a number of people. In a report edited by Daniels (5), the results of the transport of tritiated water and chlorine-36 away from the Cambric event to the satellite well at RNM-2S were compared with the two-dimensional calculations of Sauty (12). At that time, curves based on a Peclet number of 10 gave excellent fits to the data through the maxima, but they also indicated that the data would diverge from the

curves for both tritium and chlorine-36 at longer elution times. This excellent fit, however, could only be achieved by allowing the tritium source term to float; the best fit was a value that was 70% of the calculated source term. Travis, in the same report (5), made a three-dimensional numerical simulation of the transport at Cambric using the TRACR3D model (13). Both the leading edge of the elution front and the time of the appearance of the peak concentration of tritium agreed well with the observed data . There were insufficient results at that time to model the tail of the elution curve.

With the addition of data from 1983 to the present, the results have been reexamined using the Sauty (12) model. Although a curve with Peclet number of 10 (Figure 3) fits the tritium data through the maximum, now, a better fit to the tailing portion of the data can be made using a curve with Peclet number of 6 (Figure 4). The solution for chlorine-36 calculated with a Peclet number of 6 is shown in Figure 5. However, a difference in slope between the data and the Sauty curve at longer pumping times is still evident for both tritium and chlorine-36 and may indicate that a curve with still lower Peclet number is needed.

The dimensionless time (T_r) of the Sauty curves , given in the upper abscissa in Figures 3-5, equals 1 for the time required to pump the volume of water contained in a cylinder whose radius is equal to the distance from the source term to the well. The volume of water from which the chloride ion is excluded is the difference in volumes for $T_r=1$ on the chlorine-36 curve (6.5×10^6 m^3) and on the tritium curve (8.6×10^6 m^3). From these values the chloride exclusion volume is 2.1×10^6 m^3. The mass of the alluvium contained in the same cylinder can be estimated from some of the known or estimated properties of alluvium. The bulk density of the alluvium (1.46 g/cm^3) is calculated from the grain density (2.51 g/cm^3) determined on a side-wall sample taken from 260 m in RNM-1. This number, combined with an estimated porosity (~0.42 total) for inter and intragranular porosity (5), yields a mass of alluvium of 2.43×10^{10} kg in this cylinder. Anion exclusion, by convention, is listed as milliliters of liquid excluded by 100 g of soil. Thus the calculated

anion exclusion of the alluvium at the NTS is 8.6 ml/100 g. Previously, the anion exclusion was calculated to be 5.0 ml/100 g using a Peclet number of 10 in the Sauty curves (5). Thomas and Swoboda (11) found anion exclusion for various soils to vary from 3.7 ml/100 g for sandy loam soil to 20 ml/100 g for clay that was 40% montmorillonite. An alluvium had an anion exclusion of 3.9 ml/100 g, a value somewhat lower but comparable to the ones we calculate.

A Peclet number of 6 corresponds to a dispersivity of 15.1 m for the geometry of this experiment, whereas a Peclet number of 10 corresponds to a dispersivity of 9.1 m. These values can be compared to the longitudinal dispersivities reported by Borg et al. (14), which were estimated by calibrating mathematical models of transport against observed transport in field studies. Dispersivities were in the range of 11.6 to 91 m for a wide variety of lithologies. A sand or gravel deposit, which probably most closely approximates the tuffaceous alluvium of the NTS, had a calculated dispersivity of 21.3 m.

It appears that as pumping is continued with time, a Sauty curve with a lower Peclet number best fits the latest portion of the elution data. The calculated dispersivity, correspondingly, increases with time of pumping—an effect that has sometimes been called a "scaling effect" of dispersivities in field experiments. Matheron and de Marsily (15) propose that the scaling effect observed in field experiments is an artifact of the model and that the convection-diffusion equation will not fit the transport of solutes except for large distances or long times because diffusion behavior is not homogeneous in three dimensions. They suggest that all media are layered to some extent and that the different dispersivities of each of the horizontal layers as well as the vertical velocities must be considered.

The present field experiment presents an opportunity to model the transport of solutes in a relatively homogeneous–but layered media. Methods are being developed to calculate dispersivities as a function of concentration of the solute on the Sauty curve at different times. Such calculations will show if an asymptotic macrodispersion of Matheron and de Marsily (15) is ever reached as the experiment is continued. An additional important

benefit will be obtained by continuing this experiment until it is successfully modeled; it can then be used for validating codes to be used in the calculations of transport away from a high-level nuclear waste repository.

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CAPTIONS

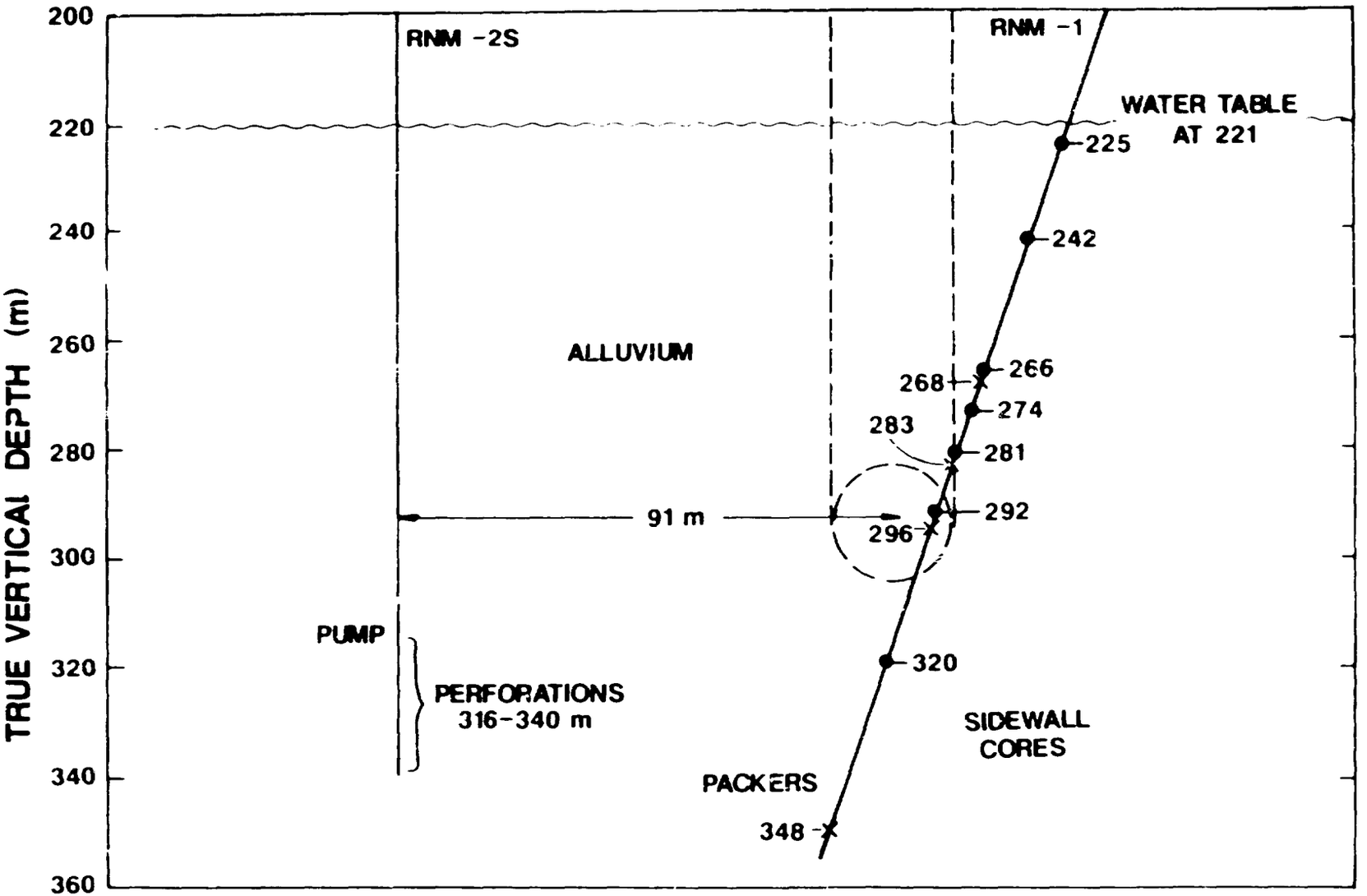
Figure 1 Cambric Single-Well Pumping Test

Figure 2 Chlorine-36 and Tritium Elution Curves

Figure 3 Tritium Elution Data Compared with a Sauty Curve (Peclet Number Equal to 10)

Figure 4 Tritium Elution Data Compared with a Sauty Curve (Peclet Number Equal to 6)

Figure 5 Chlorine-36 Elution Data Compared with a Sauty Curve (Peclet Number Equal to 6)


TRUE VERTICAL DEPTH (m)
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220
240
260
280
300
320
340
360
RNM -2S
RNM -1
WATER TABLE
AT 221
225
242
ALLUVIUM
266
268
274
283
281
91 m
292
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PUMP
320
PERFORATIONS
316-340 m
SIDEWALL
CORES
PACKERS
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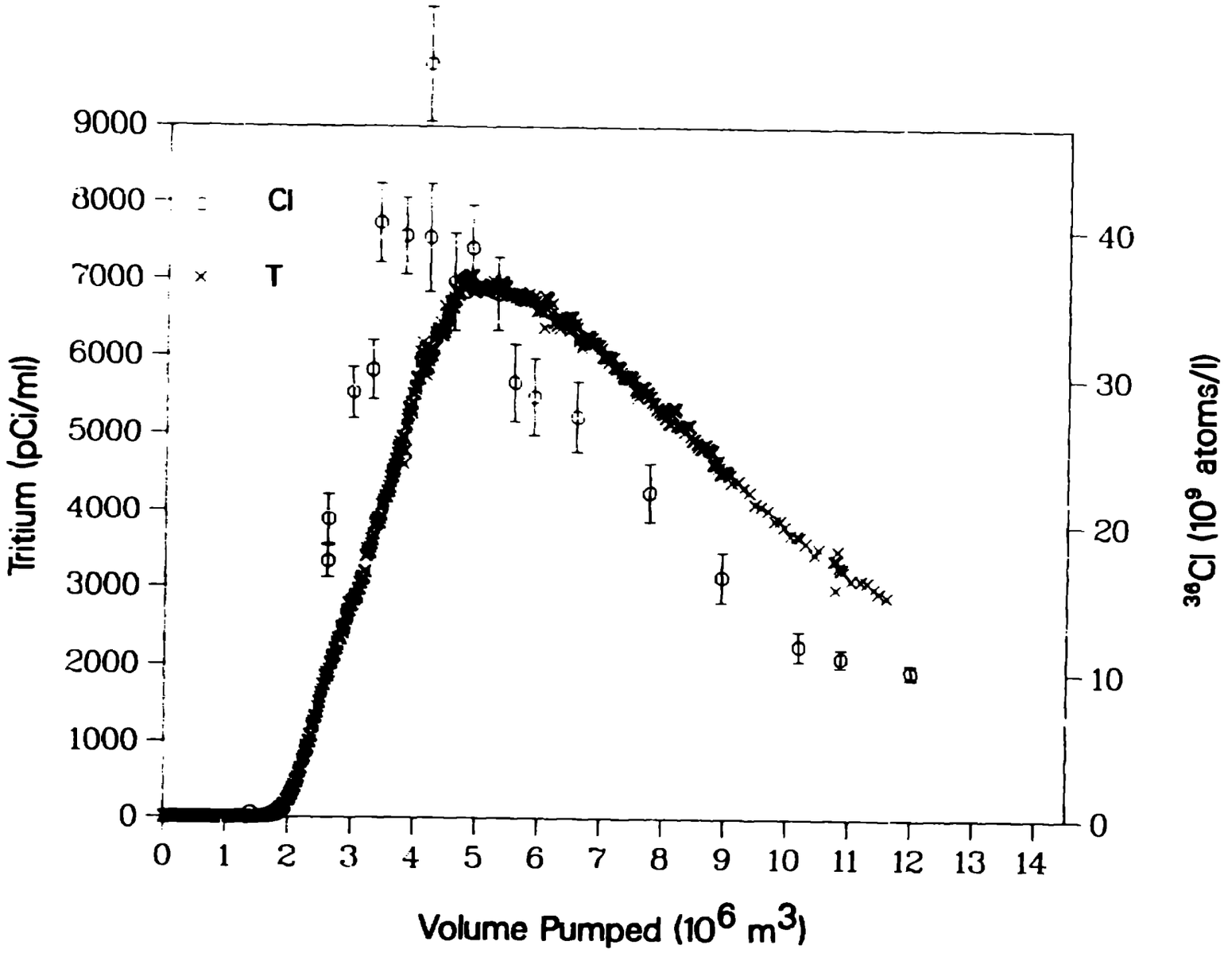
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7000
6000
5000
4000
3000
2000
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^{36}Cl (10^9 atoms/l)
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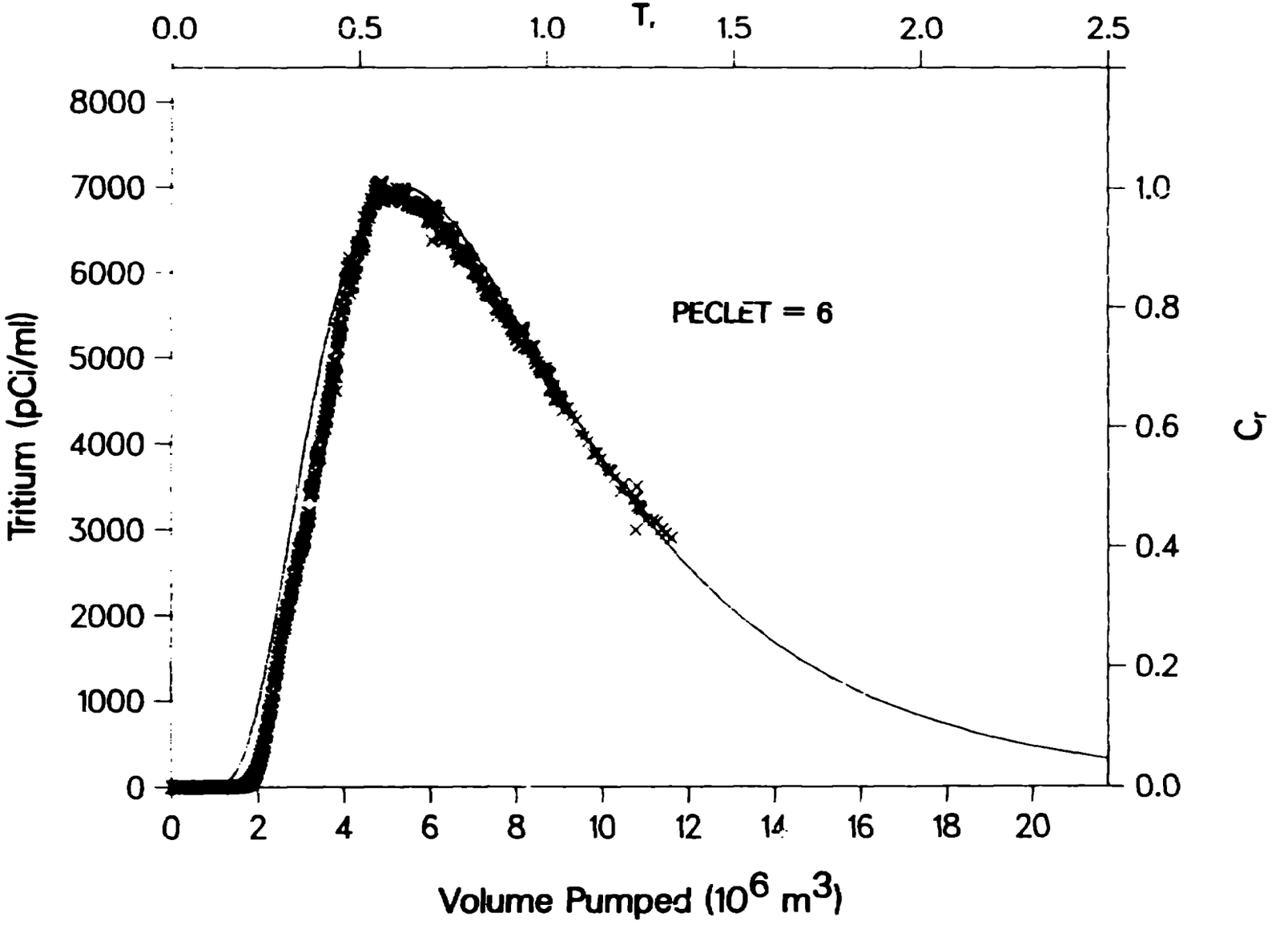
Volume Pumped (10^6 m^3)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1.5
2.0
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7000
6000
5000
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2000
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0
Tritium (pCi/ml)
PECLET = 6
1.0
0.8
0.6
0.4
0.2
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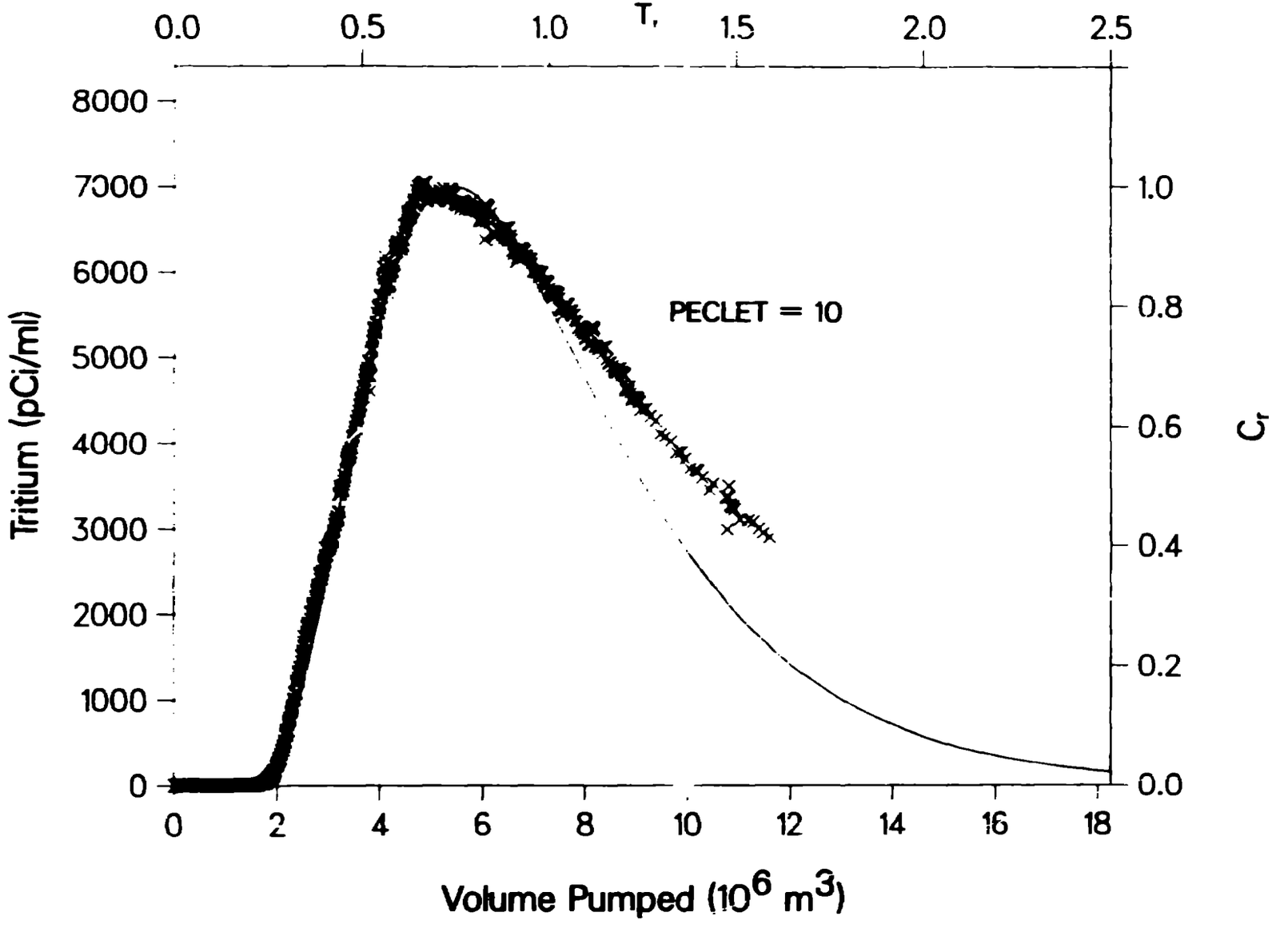
C
0
2
4
6
8
10
12
14
16
18
20
Volume Pumped (10^6 m^3)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1.5
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Tritium (pCi/ml)
PECLET = 10
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0.6
0.4
0.2
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8
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16
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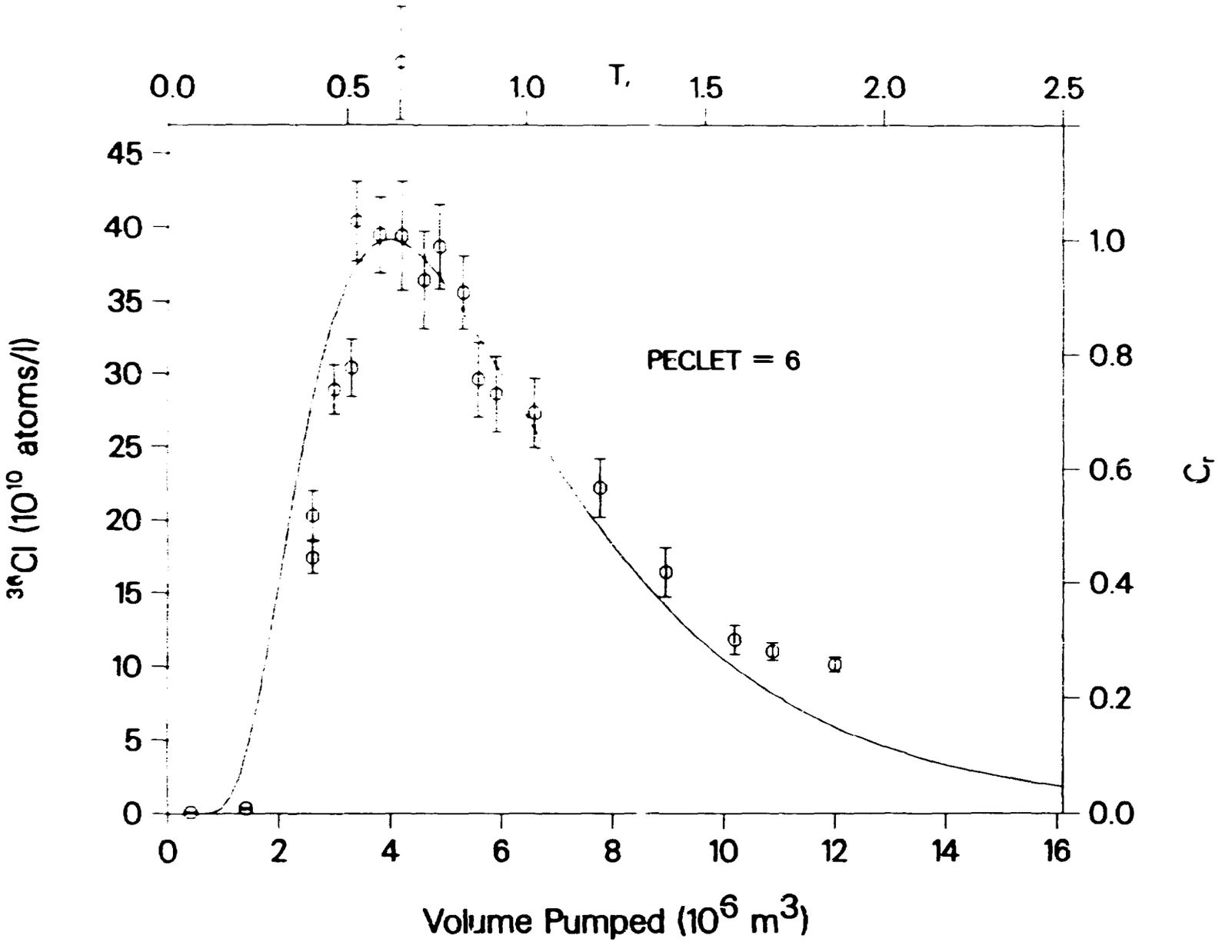
Volume Pumped (10^6 m^3)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T,
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2.5
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35
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25
20
15
10
5
0
PECLET = 6
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0.8
0.6
0.4
0.2
0.0
C/C
^{36}Cl (10^{10} atoms/l)
0
2
4
6
8
10
12
14
16
Volume Pumped (10^6 m^3)