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Radioactive Composition of Airborne Particulates at Ambrosia Lake, New Mexico

by

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Abstract

The concentration and distribution of radionuclides in airborne particulates from the Ambrosia Lake Uranium mining district in New Mexico are discussed.

Analysis of over 500 weekly samples, collected over four years at eight various locations including the Kerr-McGee uranium mill area, has provided a large data-base.

A description of the airborne radiological environment in the district resulting from the chemical and statistical evaluations of the data-base is presented.

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INTRODUCTION

Kerr-McGee commenced sampling ambient airborne particulates at eight selected locations near its Ambrosia Lake mill in 1977. The purpose of the sampling was to characterize any regional radiological environmental influences from mill operations, ore storage or ore haulage.

From 1977 through 1981, over 500 samples, including both individual weekly samples, and composite samples, were taken and analyzed. The raw data was sufficiently complex that mathematical treatment of the data was used to provide meaningful interpretation. From an analysis of the data, specific conclusions on the ambient airborne radionuclide particulates were determined by comparison to characteristic radionuclide distributions for naturally occuring weathered outcrops, mined ore, and yellowcake.

EXPERIMENTAL AND RESULTS

Four particulate samplers were purchased and located at four points. Subsequently, some samplers were moved to other locations, as deemed appropriate, to secure representative samples near the mill and mining activity areas and at isolated background sites. The sample locations are noted on the Ambrosia Lake area map provided as Figure 1. Prevailing wind patterns, typically out of the west-north-west, were used to establish several of the sampling points.

Hi-Vol samplers (EPA), equipped with plastic filters, were used for obtaining the weekly 48-hour samples. The Hi-Vol samplers were installed with flow controllers in order to maintain a constant flow rate (1.4 m^3/min.) throughout the sampling period. The samples typically were collected over a period of two days each week and were then forwarded to the Kerr-McGee Technical Center in Oklahoma City for analysis. Upon receipt, the sample filters were ashed* and the resulting ash analyzed for the uranium decay series radionuclides (U-235, Th-234, Th-230, Ra-226, Pb-210) by gamma spectrometry and later during the program, for Be-7.

Upon completion of analyses for a calendar quarter's individual weekly samples, the samples were combined into a quarterly composite, according to location and Th-234, Pb-210 and Be-7 remeasured by a Ge(Li) gamma spectrometer. The composite was then dissolved and the radioactive constituents chemically separated; the U-238 was determined chemically (fluorometric), the Th-230 was determined by alpha spectrometry, and the Ra-226 was determined by alpha counting. The mean radionuclide concentrations in $pCi/10^6$ l for each of the site locations are given in Table 1).

*Dry ash at 550°C in O_2 - atmosphere results in no detectable loss of radionuclides.

A common method of examining data of this type and quantity is to attempt a correlation to observe whether the data points may be described using a standard distribution function. As is typical of many natural phenomena, the data were determined to fit a log-normal distribution.[1,2] The data, arranged in ascending concentrations for each isotope and location, were plotted with the abscissa (y-axis) being the logarithm of the concentration in picocuries per 1,000 cubic meters ($pCi/10^6 l$). The graph ordinate (x-axis) consisted of a probability scale commencing at 2% and terminating at 98%. These results are given in Figures 2 through 10 for all data, according to location.

In applying and interpreting the data, it must be noted that the analysis of an isotope is limited by its detection level, which is somewhat dependent upon both background interferences and activity in the sample from other nuclides. Therefore, the data points reported as "less than" were assigned a value 50% of the "less than" value with a line representing $\pm$100% of the plotted value indicating the range. Such treatment assumes that the analysis report of a "less than" value actually indicates the upper limit of the range; therefore, the value was located at the midpoint. This midpoint value then served as that discrete sample data point in all subsequent mathematical treatment. Where a composite was reported as "less than", the composite had been analyzed by gamma spectrograph; subsequently chemical separation for detailed analysis was conducted as previously described to provide a discrete data point.

Associated with the radionuclides occurring naturally in the particulates native to the region, are interferences from the presence of background radiation naturally occuring in the form of stratospheric radionuclides, such as Be-7, and certain fallout debris resulting from atmospheric nuclear testing conducted by foreign nations preceeding and during the period. Beryllium-7 is formed from spallation on Oxygen-16 and Nitrogen-14 as a result of cosmic bombardment. See NCRP Report No. 45[3] for a discussion of this phenomenon.

In addition to the analysis of airborne particulates, Kerr-McGee collected samples of the various uranium bearing materials and wastes that could also be a source of radioactive airborne particulates. The typical radionuclide distribution of these materials is provided in Table 2.

DISCUSSION

Statistical treatment of the mean radionuclide concentrations for each site location using the Student's t-test indicated that the eight sample locations and composite are essentially indistinguishable from one another. They also show patterns typical of the radionuclide disequilibrium for windblown weathered natural rock (Table 2) and sand in the locale. This indicates the greatest contribution to the measured airborne radioactivity was from the general area background sources, rather than from the ore, tailings and yellowcake.

Background locations for comparison purposes and study of the milling facility contribution were based at Thoreau and at Section 35/36.[4,5,6] sources, rather than from the ore, tailings and yellowcake. The only noted deviation from background levels occurred in samples that were obtained where uranium ore was close. For example, the schoolhouse is located close to the Sec 30 and 30W mines, where ore is stored on pads. Section 35/36 lies five miles east south east (downwind) of the K-M uranium mill. This sampler location was chosen because it would offer a good check on the dispersion of any mill airborne particulate discharges detected by the close-in samplers. Additionally, the dispersion models of other investigators supported the choice of this location as a background zone.[4,5,6,7] Thoreau lies approximately 28 miles south south west of Ambrosia Lake along Interstate Highway 40. Thoreau is sufficiently remote from Ambrosia Lake that it should not be influenced by the minerology, mining, or milling activities at Ambrosia Lake. The experimental results support the choice of these two locations for background sampling (Table 1 and Figures 2 and 3). It should be noted that our results for Thoreau are similar to the results of Momenti and Kisieleski for their background Location.[5]

The secular disequilibrium noted in the data from the particulate sampling stations listed in Table 1 compares closely with the distribution for the weathered ore/outcrops noted in Table 2. Disequilibrium in both the air samples and weathered materials is restricted to the suppression of radium which, being somewhat water soluble, has apparently been leached by natural weathering. This degree of radium disequilibrium is much greater than that observed for the -325 mesh fraction of native uranium ore, which shows a much smaller deviation from equilibrium for the radionuclides of interest.

The radionuclide disequilibrium observed from the air particulate samples was totally different than that indicative of either mill tailings or yellowcake (See Table 2). Tailings are greatly enriched in radium and somewhat enriched in thorium over uranium, whereas yellowcake is greatly enriched in uranium over the other species.

The schoolhouse location is situated in close proximity to the uranium ore storage and loading pads at Mines 30 and 30W. The air particulate data for that station suggests a minor contribution from ore dust. This site impact is postulated based on the prevailing wind direction and the higher Ra226/U238 ratio than that noted for the other sites which are more typical of weathered species (see Table 2). The high degree of correlation between the various sampling points showing the airborne radioactivity to be from indigenous weathered material rather than from specific mining and milling activity, is enlightening. It is interesting to note that similar findings showing

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*The term "schoolhouse" refers to the building leased from the local school district, used by Kerr-McGee's Health Physics and Environmental group from May, 1976 until November, 1982. The building had not been used for classroom instruction since 1969.

a rather short range influence of radon emmanation from tailings piles[7] have been reported. The radon activity rapidly became indistinguishable from natural background variances endemic to the area within a distance of a quarter mile.

CONCLUSIONS

On the basis of the above data, several conclusions have been reached:

- Comparison of the U-238 and Ra-226 data demonstrate that the radium content has been significantly depleted below secular equilibrium. This disequilibrium is not consistent with particulates characteristic of uranium milling operations or uranium ore. The radium depletion found in the Ambrosia Lake air particulates is consistent with that found in natural deposits of uranium exposed to weathering. Radium, a class IIA alkaline earth element, is solubilized when exposed to a mildly acidic solvent such as rainwater containing absorbed CO_2.

- The slope of the curve for Be-7 concentration indicates that this isotope, resulting from spallation of N-14 and O-16 by cosmic radiation, represents a single source of origin, i.e. the stratosphere.

- Conversely, the steepness of the other curves suggest multiple sources for the air particulates. The sources are not homogeneous and may or may not include material originating from technically enhanced sources.

- The Pb-210 curve is distorted upwards at higher concentrations. Since Pb-210 results from Rn-222 decay, it is believed that this isotope (Pb-210) accompanies Be-7 as it is injected into the troposphere by meteorological events.

- The radionuclide composition of the air particulates in the Ambrosia Lake area suggests that the overwhelming majority of the particulates originate from natural background sources.

- A sampling station location may show radionuclide compositions influenced by a nearby source of radiaoactive particulates. The isotopic analysis of the possible source will confirm the contribution to background.

- In the future, analysis should only be performed on composite quarterly samples due to the variability in the natural sources of individual weekly samples.

References:

1. Aitchison, J. and J.A.C. Brown, The Long-Normal Distribution, Cambridge University Press, Cambridge, England, 1957.

2. Waite, D.A., and D.H. Denham, "Identification of Potential Improvements in Environmental Surveillance Techniques", Nuclear Safety, 17, 344, 1976.

3. NCRP Report No. 45, Natural Background Radiation in the United States; November 15, 1975.

4. ANL-8007: "Environmental Monitoring at Argonne National Laboratory", Annual Report for 1972.

5. NUREG/CR-1320, Measured Concentrations of Radioactive Particulates in Air in the Vicinity of the Anaconda Uranium Mill, February 1980.

6. NUREG/CR-1407, A Field and Modeling Study of Windblown Particulates from a Uranium Mill Tailings Pile, June 1980.

7. Shearer, S.D. Jr., and C. Sill, "Evaluation of Atmospheric Radon in the Vicinity of Uranium Mill Tailings", Health Physics, 17, 77, 1969.

TABLE 1

MEAN* AIRBORNE RADIONUCLIDE CONCENTRATIONS

($pCi/10^6 l$)

LOCATION	NO. OF SAMPLES	U 238	Th 234	Th 230	Ra 226	Pb 210	Be 7
COMPOSITE	547	5.71 ± 13.77	3.12 ± 15.0	5.64 ± 6.90	0.88 ± 21.27	9.01 ± 12.0	107 ± 5
Sec 35/36	48	2.91 ± 8.70	1.08 ± 10.29	6.72 ± 6.99	0.19 ± 11.97	8.10 ± 9.03	94.2 ± 7.5
THOREAU	50	4.24 ± 10.62	2.31 ± 13.86	4.10 ± 5.85	0.18 ± 16.59	3.75 ± 9.96	--
SCHOOLHOUSE	145	7.53 ± 12.90	5.99 ± 14.04	7.10 ± 6.96	3.25 ± 46.83	12.6 ± 10.5	114 ± 5
SUBSTATION	57	3.42 ± 12.33	2.41 ± 11.16	5.95 ± 5.94	0.26 ± 9.84	21.3 ± 9.1	111 ± 5
MILL SWAMP	57	3.95 ± 10.77	2.03 ± 11.7	6.37 ± 6.54	0.78 ± 15.45	22.6 ± 7.6	101 ± 5
TRAILER	104	7.10 ± 13.53	6.06 ± 12.75	4.18 ± 6.99	1.84 ± 9.54	4.77 ± 14.07	109 ± 6
DECANT POND	68	9.17 ± 18.30	1.58 ± 15.0	5.52 ± 6.99	0.79 ± 27.42	6.25 ± 9.87	116 ± 5
Sec 24	18	6.49 ± 31.11	22.4 ± 40.2	4.72 ± 7.56	0.16 ± 20.37	5.18 + 11.64	--

* Mean ± 3

TABLE 2
ANALYSIS OF URANIUM BEARING NATURAL MATERIALS,
TAILINGS, AND YELLOWCAKE IN THE AMBROSIA LAKE AREA
(pCi/gm)

	RADIONUCLIDE			
MATERIAL*	U-238	Th-230	Ra-226	Pb-210
WEATHERED ORE/ OUTCROPS				
Abandoned Mine	$1.1 \times 10^5 \pm 5 \times 10^3$	$1.2 \times 10^5 \pm 4 \times 10^2$	$6.6 \times 10^3 \pm 3 \times 10^1$	$8 \times 10^3 \pm 3 \times 10^3$
Westwater Outcrop	20 ± 2	26 ± 1	10 ± 1	4.8
URANIUM ORES				
KM Sec 30	1000 ± 50	3060 ± 20	1480 ± 70	76
KM Sec 22	1400 ± 80	1380 ± 10	835 ± 40	130
KM Sec 10 (Weathered 10 yrs)	157 ± 8	179 ± 2	86 ± 8	14
KM MILL TAILINGS				
#1	42	380	850 ± 14	130
#2	49	440	1110 ± 20	140
#3	15	132	323 ± 5	46
#4	49	430	1020 ± 20	520
#5	40	350	657 ± 12	120
KM YELLOWCAKE**				
#1	$2.72 \times 10^5 \pm 1.8 \times 10^4$	280 ± 36	3.6 ± 1.0	-
#2	$2.47 \times 10^5 \pm 1.9 \times 10^4$	124 ± 34	-8.8 ± 1.0	-

* All samples were sieved and only the -325 mesh (U.S. series) fraction used for analysis
** NUREG/CR-1216, p. 10

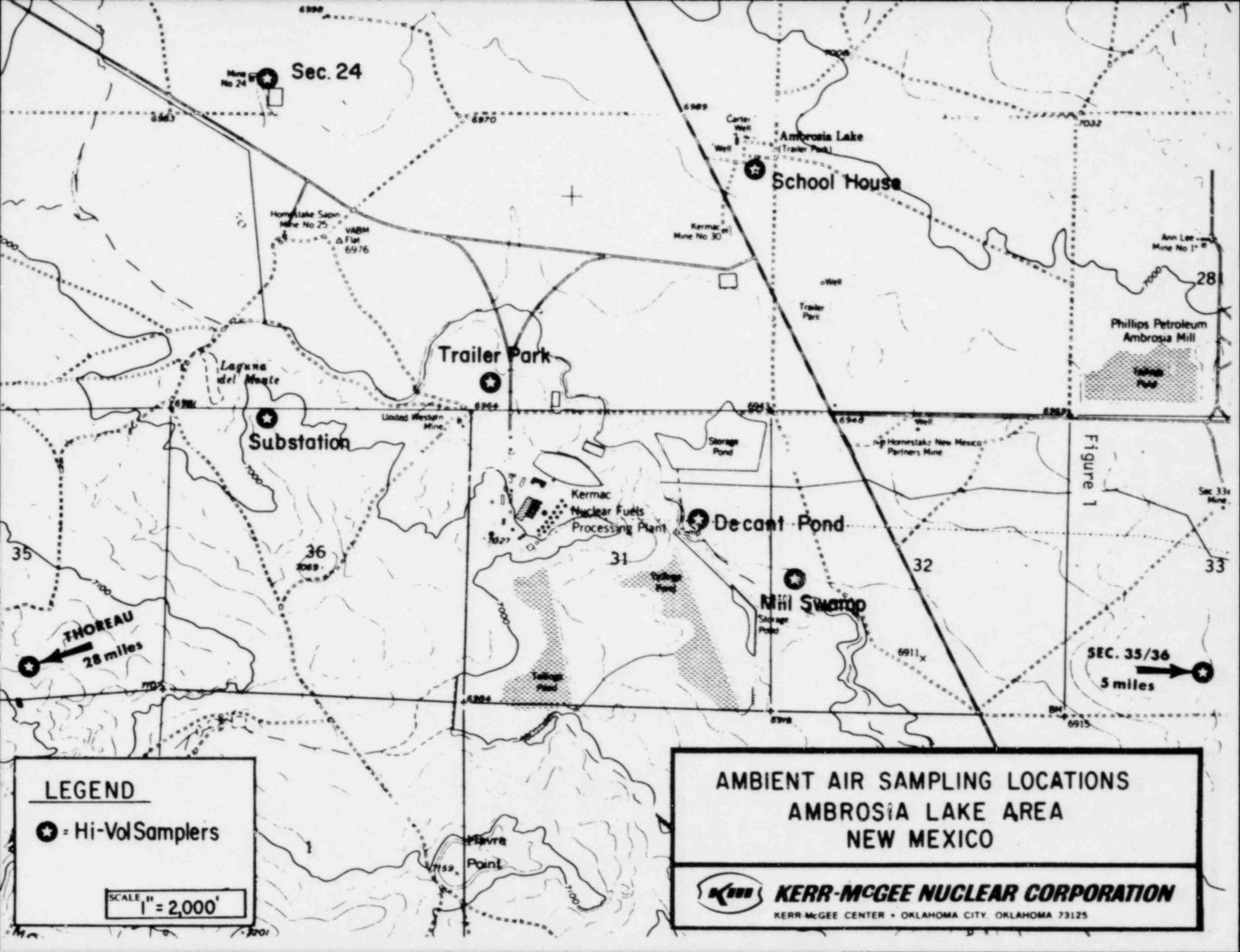
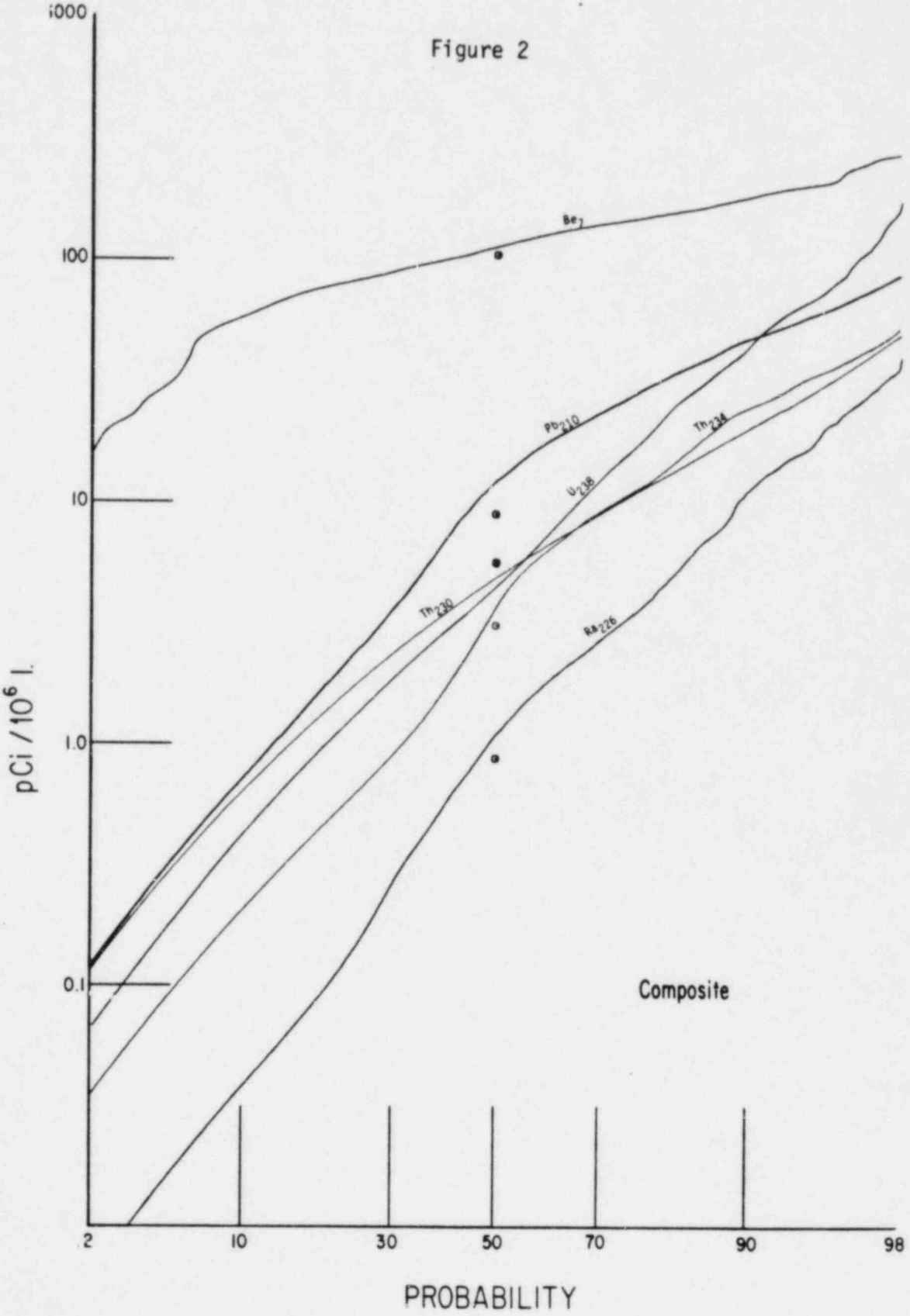

Sec. 24
Ambrosia Lake
School House
Trailer Park
Substation
Laguna del Muerto
Kermac Nuclear Fuels Processing Plant
Decant Pond
Mill Swamp
Phillips Petroleum Ambrosia Mill
Figure 1
THOREAU
28 miles
SEC. 35/36
5 miles
35
36
31
32
33
28
LEGEND
= Hi-Vol Samplers
SCALE 1" = 2,000'
AMBIENT AIR SAMPLING LOCATIONS
AMBROSIA LAKE AREA
NEW MEXICO
KERR-McGEE NUCLEAR CORPORATION
KERR-McGEE CENTER • OKLAHOMA CITY, OKLAHOMA 73125

Figure 2


1000
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10
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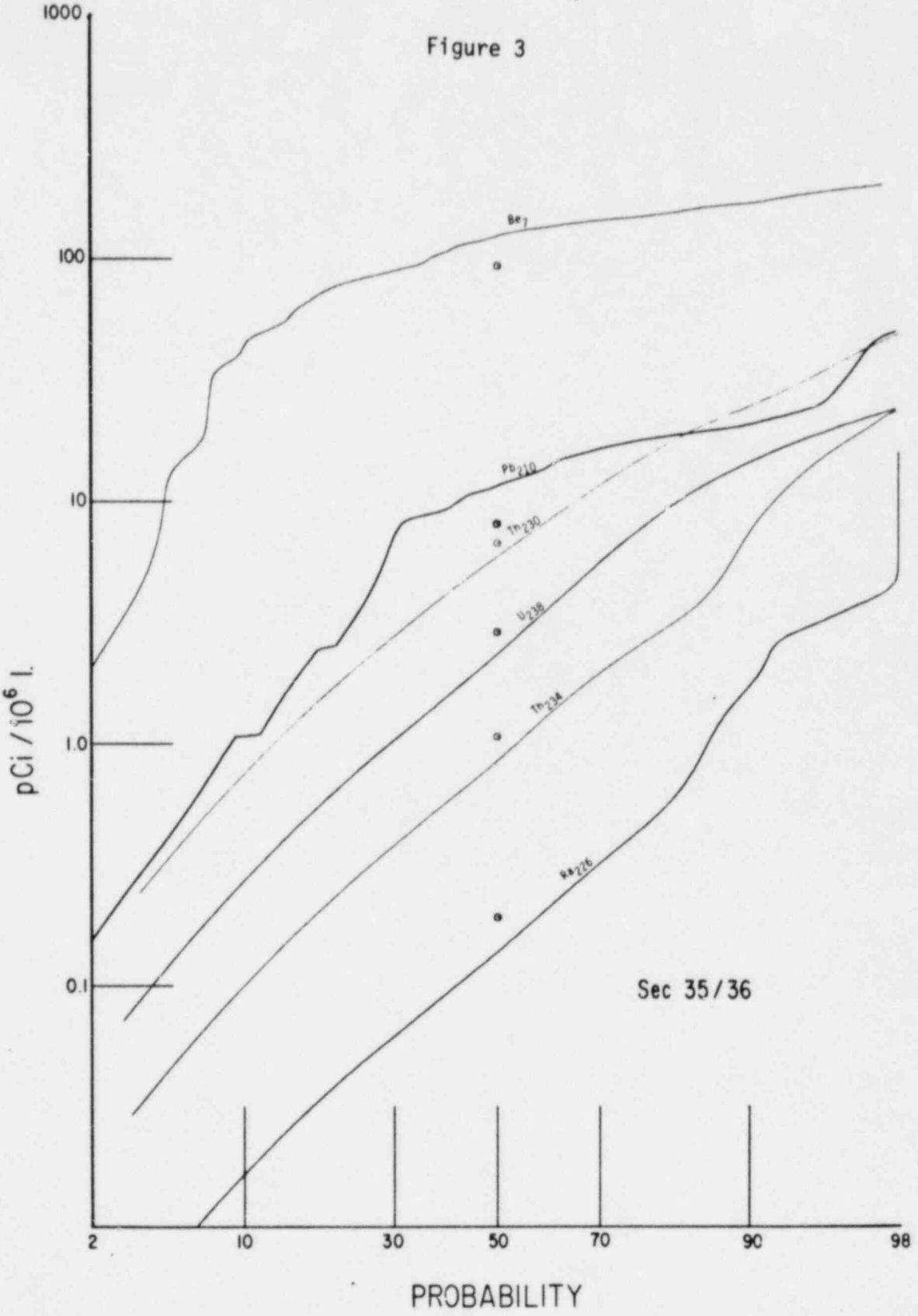
pCi / 10^6 l.
Be7
Pb210
U238
Th234
Th230
Ra226
Composite
.2
10
30
50
70
90
98
PROBABILITY

Figure 3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Be7
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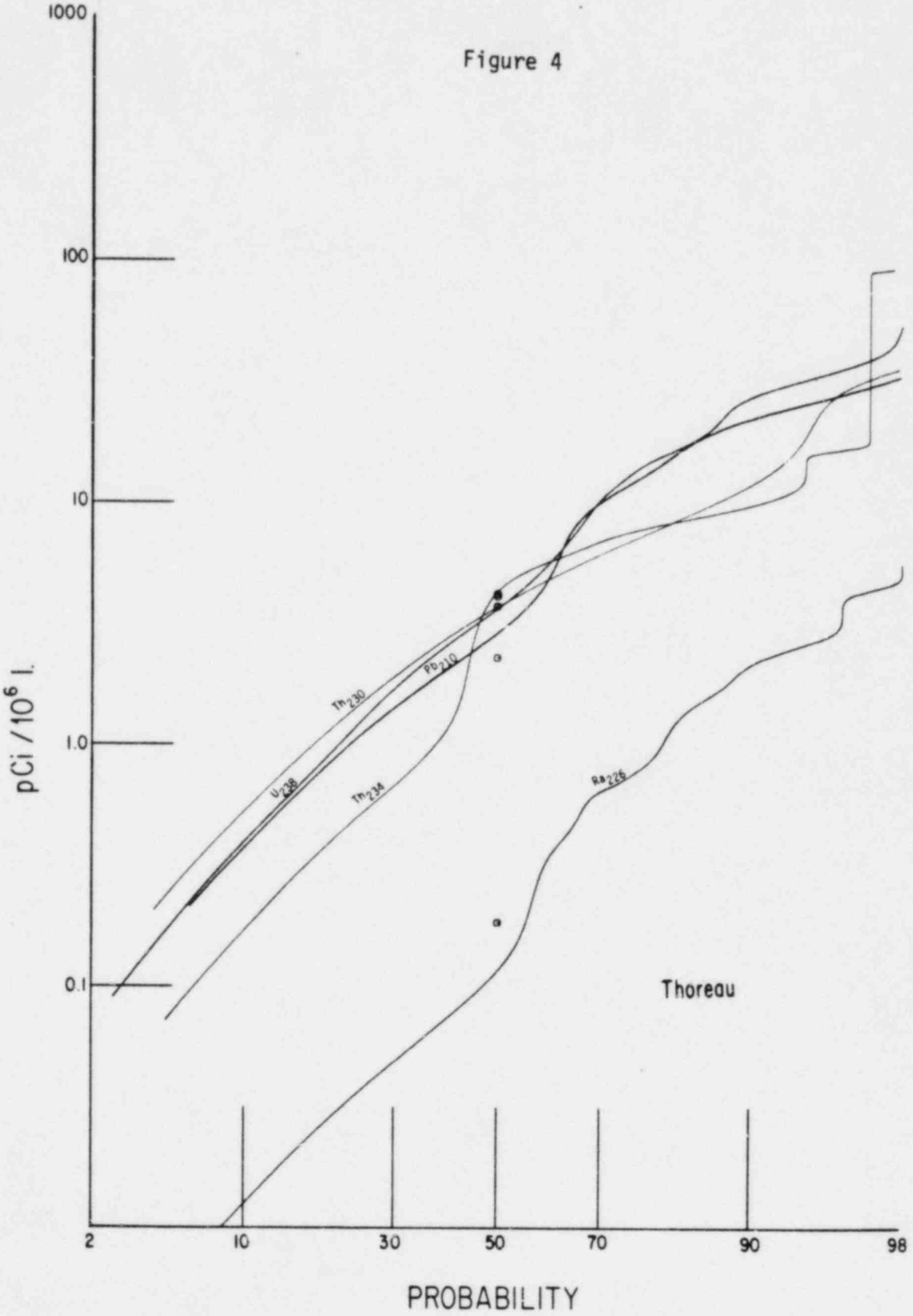
Th230
U238
Th234
Ra226
Sec 35/36
2
10
30
50
70
90
98
PROBABILITY

Figure 4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pCi / 10^6 l.
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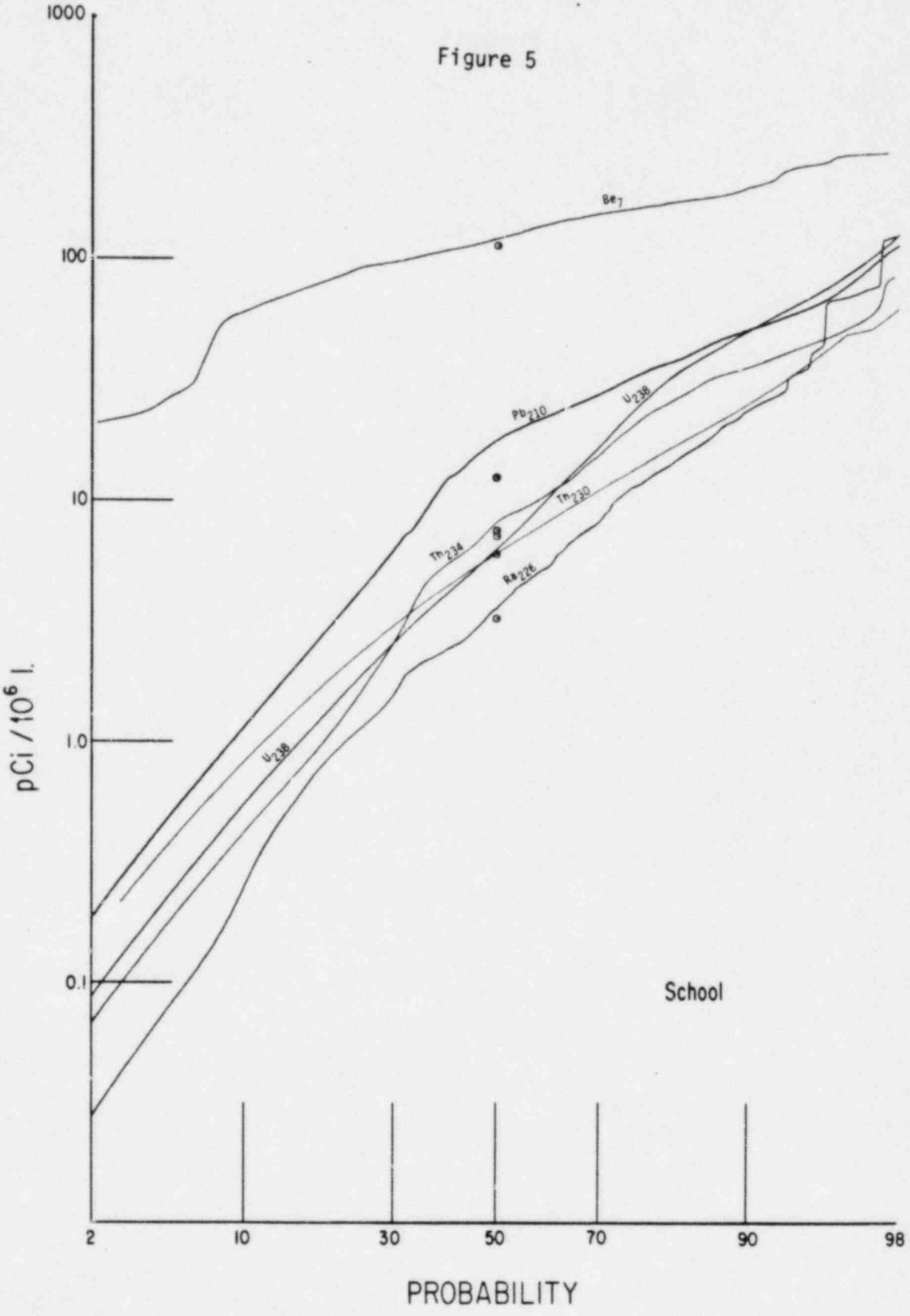
Pb210
U238
Th234
Ra226
Thoreau
2
10
30
50
70
90
98
PROBABILITY

Figure 5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pCi / 10^6 l.
Be7
Pb210
U238
Th230
Th234
Ra226
U238
School
2
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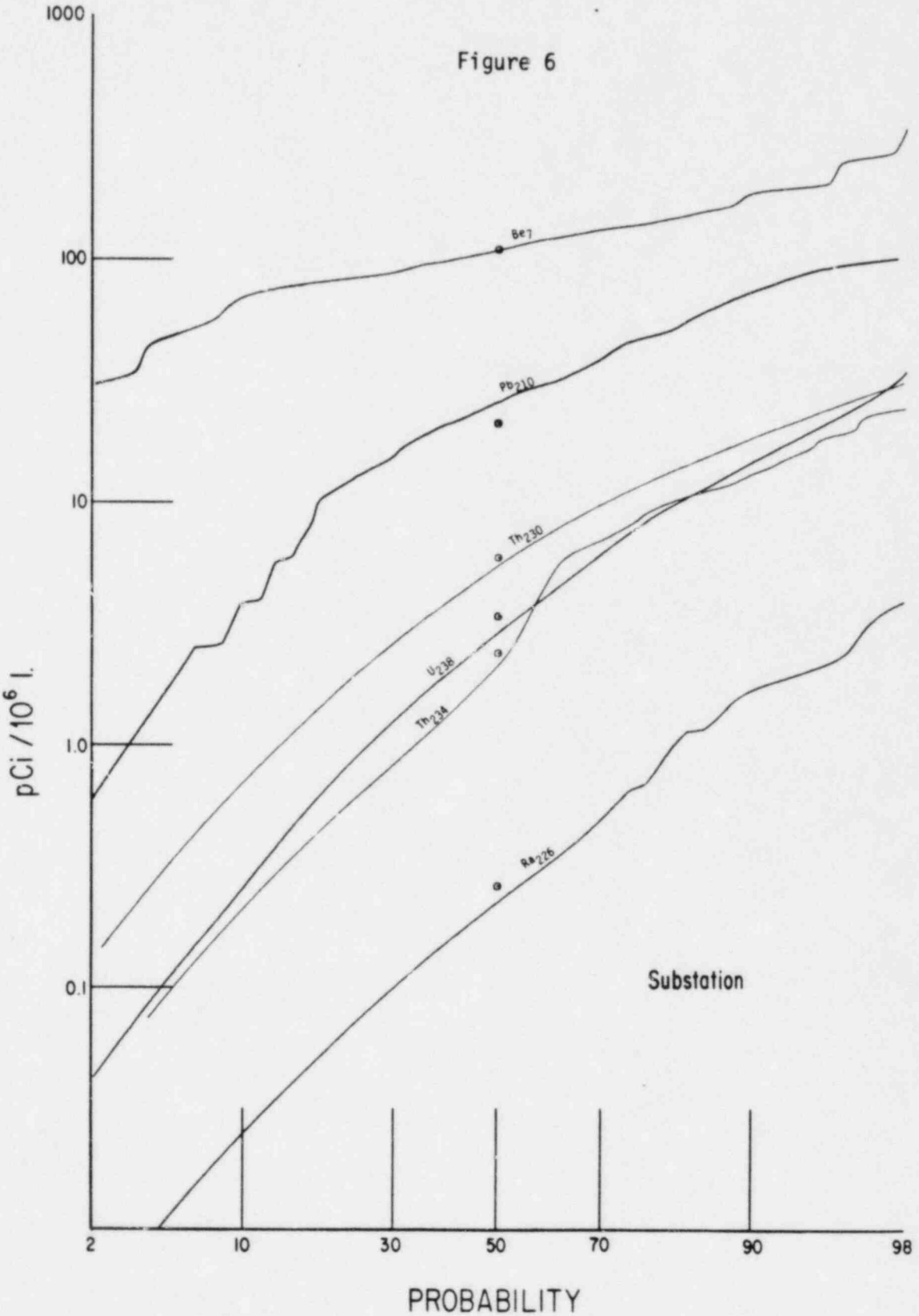
30
50
70
90
98
PROBABILITY

Figure 6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Ra226
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70
90
98
PROBABILITY

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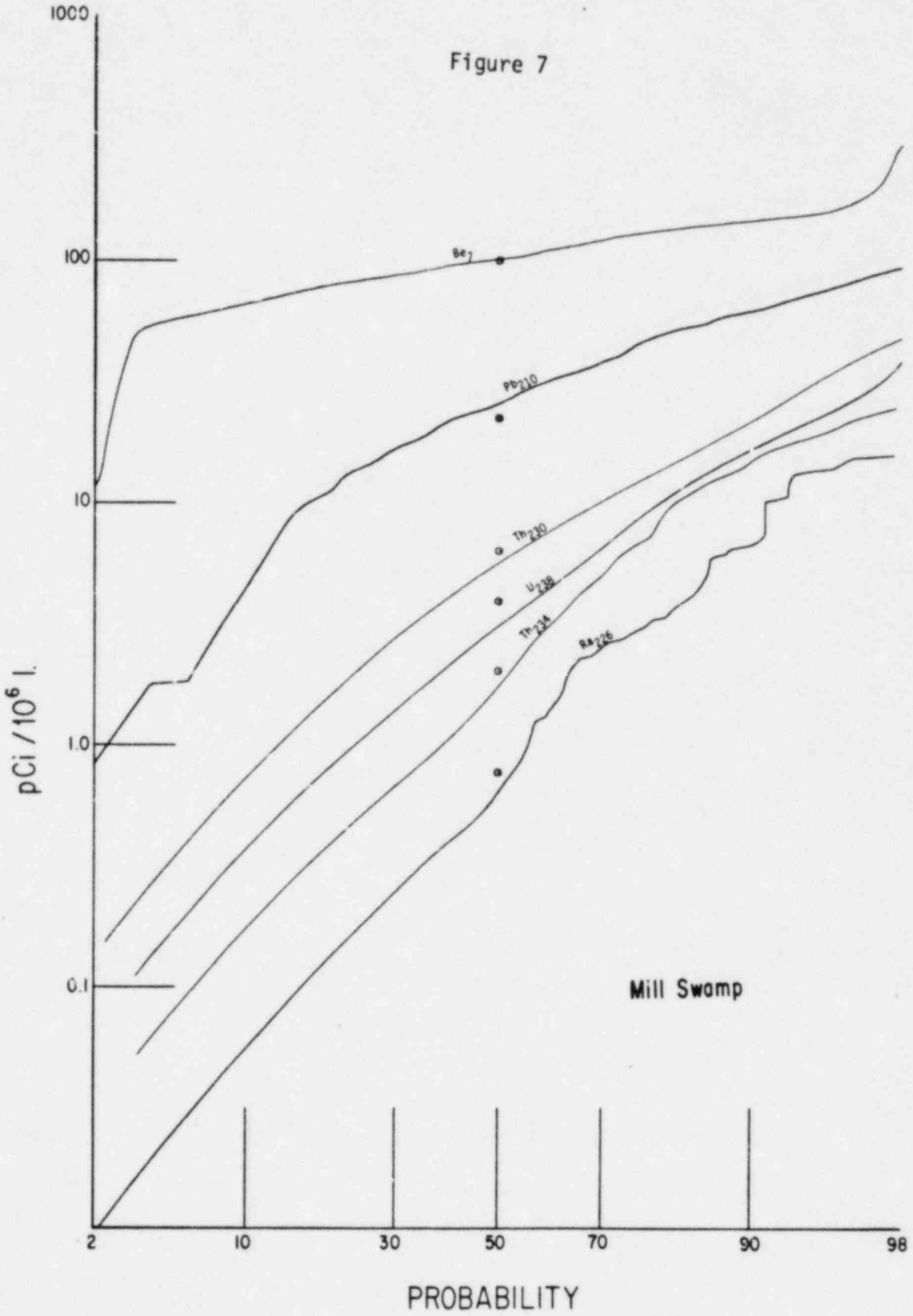
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Th230
U238
Th234
Ra226
Mill Swamp
2
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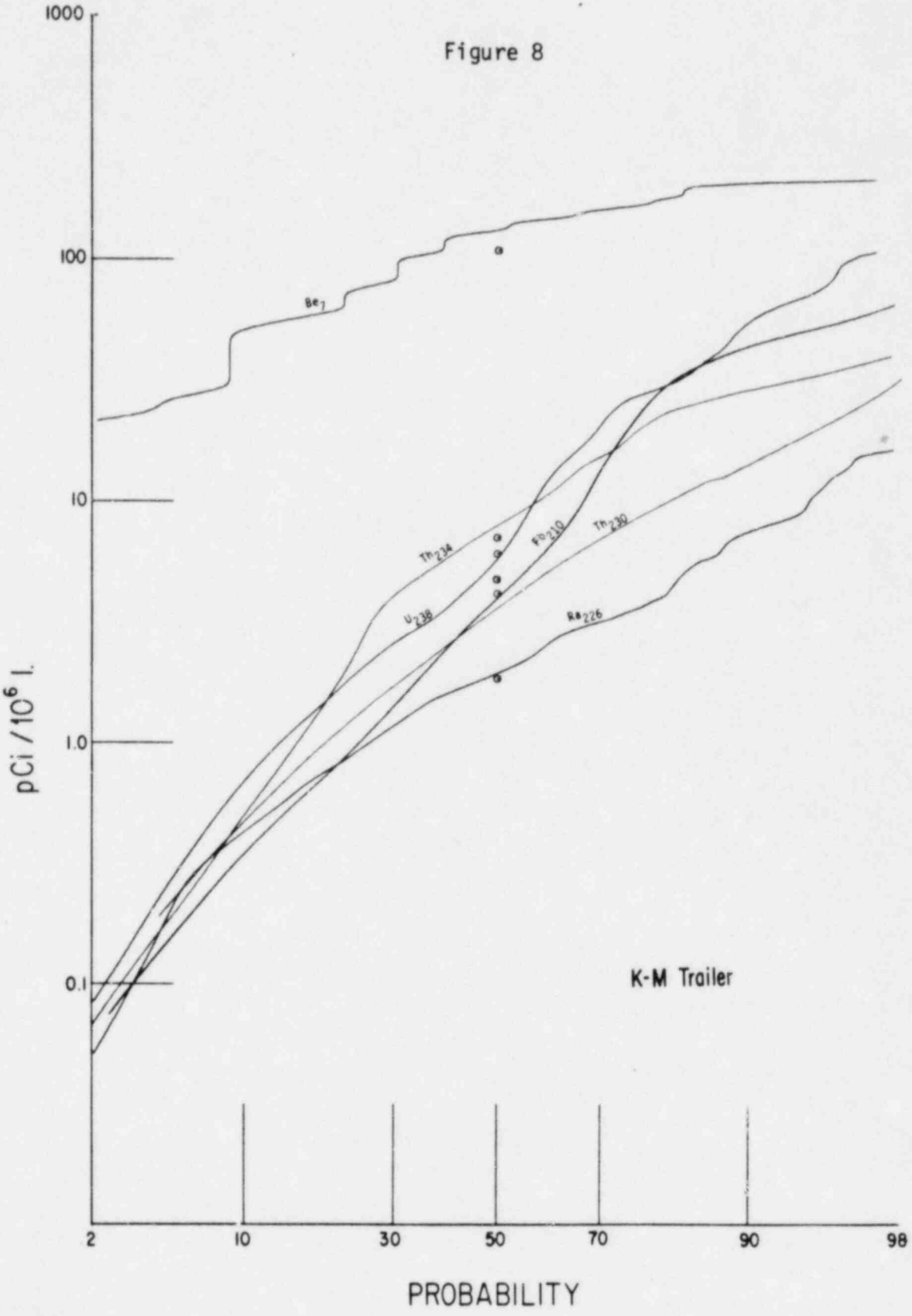
70
90
98
PROBABILITY

Figure 8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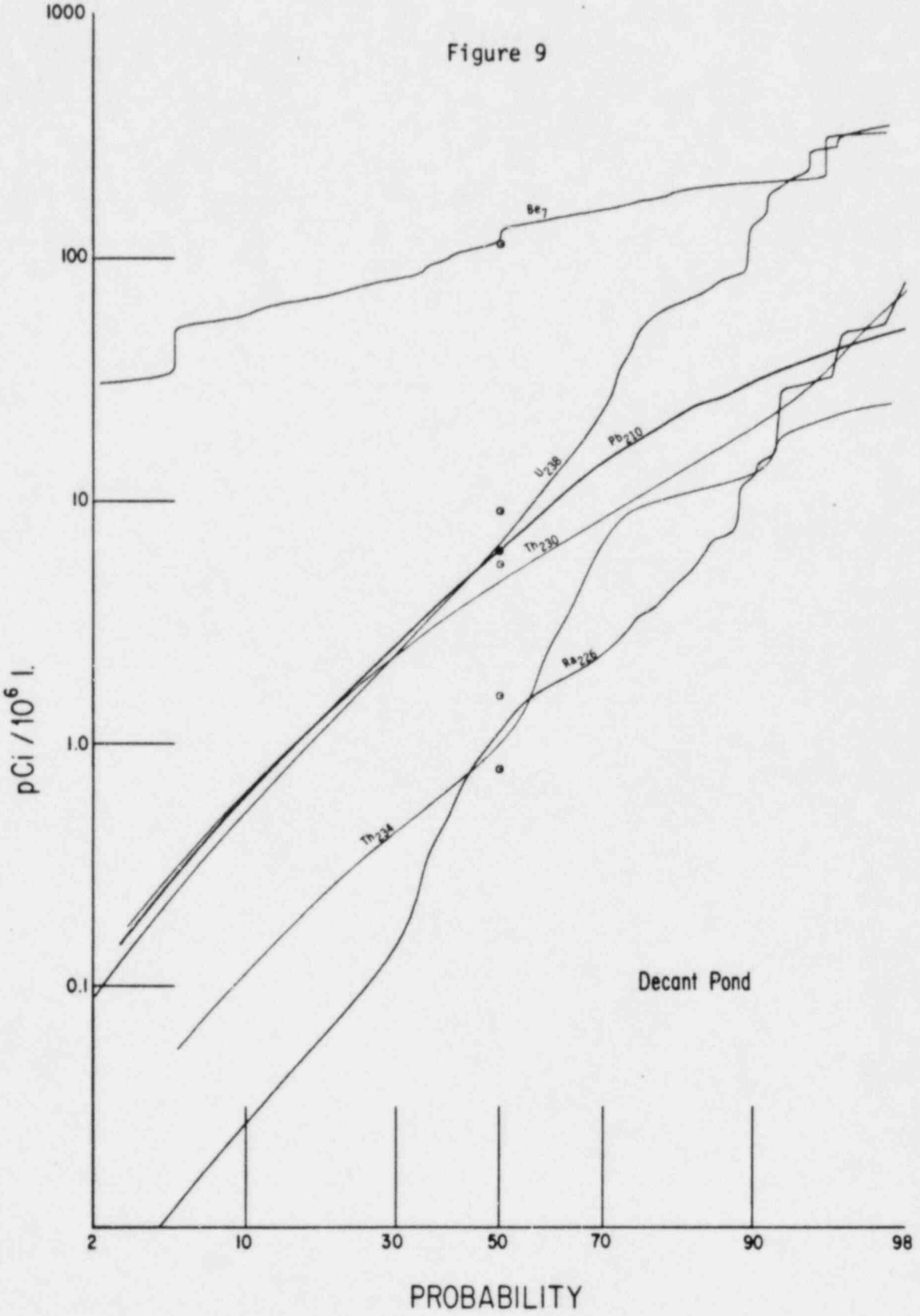
98
PROBABILITY

Figure 9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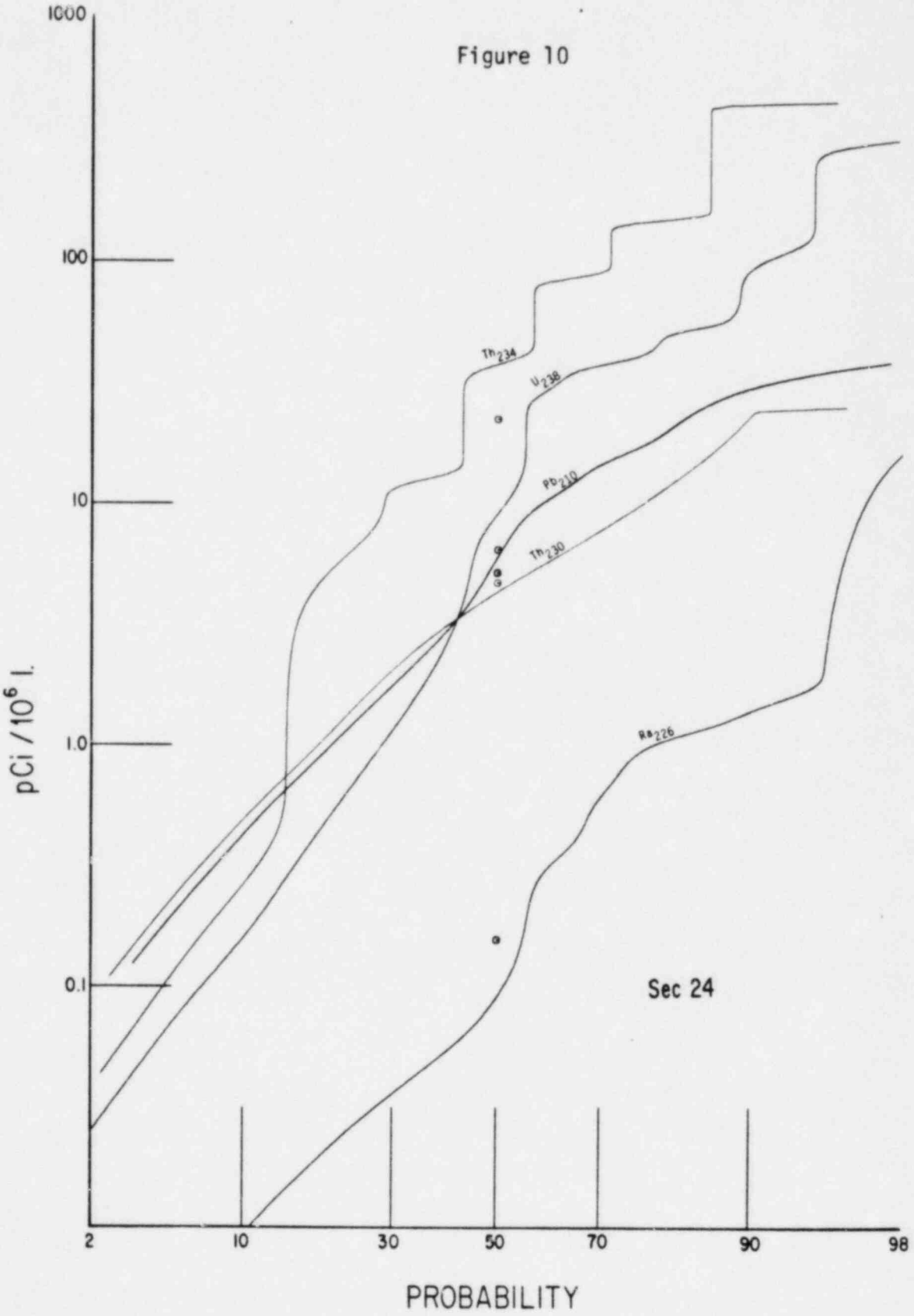
98
PROBABILITY

Figure 10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70
90
98
PROBABILITY