THE DISTRIBUTION OF PLUTONIUM AND AMERICIUM WITH DEPTH IN SOIL AT ROCKY FLATS

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ABSTRACT

In 1975, 90 percent or more of the total plutonium deposit resided in the first 10 cm from the surface at Rocky Flats. Plutonium has penetrated the soil such that the maximum concentrations no longer exist at the surface but at depths ranging from 2 to 6 cm depending upon soil conditions. Plutonium-238 follows the same patterns as Pu-239-240 while Am-241 exhibits only a slightly greater mobility. A semi-empirical model is proposed which describes the penetration of Rocky Flats plutonium with depth and also the global fallout plutonium distribution at a site in Massachusetts. This model predicts a reduction in the surface concentration of plutonium at Rocky Flats which is in agreement with the observed reduction in the mean annual air concentrations due to resuspension from the surface.

INTRODUCTION

The Rocky Flats plant which is located 25 km northwest of the center of Denver, Colorado manufactures plutonium for the production of nuclear weapons. Between 6 and 11 curies of Pu-239-240 were released to the environment when some of the barrels of contaminated cutting oil which were stored in an open field on the southeast corner of the plant leaked their contents to the ground.[1,2] Then natural air turbulence at the ground surface

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and plutonium recovery operations suspended the contaminated soil particles in the atmosphere where they were transported predominantly east-southeast by the prevailing westerly winds

The history of this contamination episode, source term, air concentrations, soil concentrations, etc , have been adequately covered in the literature [1-10] One aspect which has been only superficially studied is the penetration of the plutonium contamination into the soil.[4 5]

Penetration into soil is important because it reduces the source term available for resuspension of the plutonium into surface air. Penetration also regulates the availability of plutonium to the root zone for possible uptake in vegetation and is one of the pathways for its ultimate disposition in the environment

This report provides extensive data on concentrations of Pu-238, Pu-239-240 and Am-241 to depths of 40 cm in soil at several sites at Rocky Flats From these data we develop a penetration rate of plutonium into soil, and relate it to the observed reduction of the mean annual surface air concentration at Rocky Flats

SAMPLING

Collecting a reliable profile of samples with depth in soil is difficult for several reasons Special care must be exercised to prevent the highly radioactive surface soil from contaminating the subsurface levels during sampling. Sufficient sample must be recovered at each depth to be representative of that depth. The depth increments must be large enough that reasonable care will permit successful sampling, and small enough that important trends in the profile can be seen.

The Health and Safety Laboratory's (HASL) trench method of soil sampling[11] satisfies these criteria. In outline form, this method involves digging a trench, 1.5 m long, 0.9 m wide and 0.9 m deep. One wall of the trench is carefully shaved to make it vertical and to insure that little contamination from the surface is adhering to the wall. The vegetation at the surface of the selected trench wall is cropped closely to the surface and discarded. Samples are recovered by sequentially pressing into the prepared wall of the trench stainless steel trays of various dimensions with one edge of the trays honed to a knife edge for ease of insertion. A board is placed on the ground surface over the sampling area, and the depth below surface for each sample is measured from the bottom of the board.

In some soils and especially in many areas at Rocky Flats, stones are so plentiful and large that the insertion of trays into the trench wall is impossible. In that case, aluminum templates outlining an area of 225 [illegible] were used and the samples were recovered to the desired depths in sequence down the prepared wall of the trench with hand chisels and soil scoops. After the sample at each depth was recovered by the tray method, a buffer zone of about 7 cm on either side of the tray was cleared down to the depth of the tray to minimize debris from the upper walls of the sampling shaft falling and contaminating the lower depth samples. In the template method, this buffer zone of 7 cm extended on all sides of the template.

The smallest interval of depth that we could collect from the surface with reasonable assurance that the sample was representative of that interval was 0-2.5 cm. We devised another technique to investigate the behavior of the plutonium contamination with very small increments of depth at the surface of the soil. This technique required that the vegetation be cropped as close as possible to the surface of the soil with individual blades of grass or stems of plants being snipped just above their roots. As much as possible of the dead organic matter on the soil surface was removed and discarded without disturbing the soil surface. Then consecutive sheets of adhesive film (sticky film),

1000 cm^2 in area, were placed over the cropped area to pick up the surface particles.

As the surface particles were removed and more of the vegetative stubble exposed, careful cropping had to continue before additional sheets were used. The depth sampled by this technique is too small to be measured. Even after 27 sheets were used, we estimate the total depth sampled to be a few millimeters. However, each sheet or composite of several sheets contains debris from a slightly greater depth near the soil's surface.

The collection sites for all types of depth samples at Rocky Flats are shown in Figure 1. Site B is on the upper part of a steep slope leading south from the Rocky Flats plateau adjacent to the original barrel storage area. The vegetation was sparse at this site and the soil was dry with a high percentage of stones and rocks to a depth of about 10 cm. Below 10 cm the soil appeared to have a higher moisture content which increased with depth although the frequency of stones persisted.

Site A is further south from the plateau where the slope moderates appreciably. The vegetation was heavy in this area, and the top 10-20 cm of soil was relatively moist, contained dark organic matter and had few stones. Below 20 cm the soil was plastic and sticky. Site 7 was identified in the first HASL soil

study at Rocky Flats in 1970,[4] and is located on the relatively rich soil adjacent to Woman Creek Although the vegetation was not as thick, the soil at this site resembled the soil at Site B. However, the top layers were not as moist or did not contain as much dark organic matter.

Site C is representative of most of the contaminated region on the Rocky Flats plateau with vegetation cover, moisture and percent of stones moderate for this general area. Site D which was Site #7 in the HASL 1973 field experiment[10] is atypical of the region because 5 to 15 cm of top soil had been added in 1970 to this area for soil stabilization experiments.

Some of the sample collection techniques at Site B are illustrated in Figure 2. The radiological safety procedures were in effect because the total plutonium deposition at this site was 44,000 mCi/km^2.[10] A floorless aluminum hut was lowered onto the sampling area to minimize the amount of plutonium that would be resuspended in the atmosphere by the sampling itself. The hut was maintained at a slightly negative pressure, and all the air within the hut was filtered prior to release to the environment. These and other rad-safe precautions were provided by the Dow Chemical Company which was then the ERDA contractor at Rocky Flats.

SAMPLE PREPARATION AND ANALYSIS

The depth profile samples were air dried, weighed and pulverized in a hammer mill according to the usual HASL procedure[11] in an estimated order of low to high activity to minimize cross contamination between samples. The samples were thoroughly mixed and stored in large plastic bottles. Prior to pulverizing, most of the stones were cleaned with a brush to remove clinging soil and then discarded. This sample pretreatment was performed in a hot laboratory facility at the Brookhaven National Laboratories and made available to us by Dr. Andrew Hull of the Health Physics Division.

About 120 g aliquots of the soil samples were removed for Am-241 analysis by Ge(Li) diode spectrometry at HASL. These same aliquots were then analyzed for Pu-238 and Pu-239-240 by the Environmental Analysis Laboratories, LFE, Richmond, CA using the standard HASL leaching and analytical procedures.[11]

These procedures call for the addition of the Pu-236 tracer to the soil aliquot prior to the leaching step. Because of the high plutonium concentrations in many of these samples, large quantities of Pu-236 tracer would have had to be used. In those cases, the Pu-236 tracer was added to an appropriate aliquot of the leach solution. This modification seems acceptable because

all our previous experience with Rocky Flats soil samples indicates that the leaching removes all the plutonium

Nevertheless, to test the efficiency of leaching the soil samples in this study, LFE performed Am-241 analyses by Ge(Li) diode spectrometry on aliquots of leachates from 20 samples Since HASL had already measured the Am-241 content of the aliquot, a comparison of the two results would indicate the Am-241 recovery of the leaching procedure. We assume that this efficiency would apply equally as well to plutonium.

The sticky film samples were weighed, ashed and analyzed radiochemically by LFE for Pu-238, Pu-239-240 and Am-241. The entire ash was dissolved. The HASL procedure was used for the plutonium analysis, while the LFE procedure was used for the Am-241 analysis.[12]

As a test of the quality of the analytical results provided by LFE, blank, duplicate and reference samples were submitted blind along with the other samples in this study. The blanks were pre-1945 soil samples which contained no plutonium. The reference samples were aliquots of soil from the Brookhaven National Laboratory which was contaminated by global fallout and which had been analyzed several times to provide a reasonably known plutonium concentration.

RESULTS

That plutonium is quantitatively leached from the soil samples is inferred from the ratios of HASL's direct measurements of Am-241 in the soils to LFE's measurements of the leachates which are given in Table 1. The mean of the 19 samples (the result for S-2077 is omitted as an anomalous datum) is 1.08 ± 0.16. The 8% bias which might exist can most easily be accounted for by the imprecision of the intercalibration between the two laboratories.

The quality assurance data for the plutonium analyses are given in Table 2. The blank soil samples show no significant activity, and the reference sample results are within 5 and 13% of the expected values. One of the two sets of duplicate soil samples shows a standard deviation about the mean of ±9% or less which is within twice the standard deviations of the individual measurements. The other set of duplicate samples includes S-2077 which was shown in Table 1 to have an anomalous leaching efficiency. If an adjustment is made for this efficiency, the duplicate analyses then agree well within the standard deviations of each measurement.

The transuranic activity per gram of the sub-surface soil samples is presented in Table 3, and the areal activity of Pu-239-240 with depth is given in Table 4. Because of difficulties in sample collection, it was not always possible to get a continuous depth profile, and some slight discontinuities exist Therefore,

the Pu-239-240 mCi/km^2 cm values in column 6 of Table 3 are plotted in Figures 3, 4 and 5, and the values for the discontinuous intervals are estimated from these Figures These estimates (which are parenthetically entered in Table 4) are needed to calculate the total deposit of Pu-239-240 at each site, and the percentages of the total deposit at various depths The percent of the total deposit per cm of Pu-239-240 at each depth is given in Table 4, and is plotted as a function of depth for each site in Figures 6, 7 and 8

The transuranic activity per gram net weight of the composite sticky film samples is given in Table 5, and the nuclide ratios of both the sticky film and sub-surface soil samples are reported in Table 6.

PLUTONIUM PENETRATION INTO SOIL

It is clear from Tables 3 and 4 that measurable quantities of Pu-239-240 have been found in the soil as deeply as we have collected samples, but at 40 cm it is only 10^{-2}% of the total deposit At all three sites 90% or more of the total deposit resides in the first 10 cm from the surface.

The cumulative percent of the total Pu-239-240 deposit with depth at each site is plotted on log probability paper in Figure

9 along with the plot from less detailed depth profiles obtained earlier at Rocky Flats.[10] The data from this report do not follow a log normal distribution very well, but the results from Site 7 are in reasonable agreement with the earlier data from HASL-304 Site 7 is more similar to the sites at which the results in HASL-304 were obtained than either of the other two sites. In spite of this difference, the slope of the line for Site B in Figure 9 is similar to the slope of the HASL-304 data suggesting some similarity in the mechanisms of downward transport at all the sites at Rocky Flats. The big difference in the behavior at Site A, which will be discussed in greater detail later, is due primarily to the larger retention of the plutonium in the upper layers of soil at that site.

The reasonable agreement between the results of the cumulative deposits in this report, and total soil measurements made earlier at nearby sites provides additional verification of our data. At Site 7 where deposition gradients are shallow, 480 mCi/km^2 Pu-239-240 reported to a depth of 20 cm in 1970[4] agrees well with our estimate of 547 mCi/km^2 to that depth in Table 4.

At Site A and B, the deposition gradients are very steep, and the earlier sampling locations were not precisely surveyed. Consequently, poorer agreement should be expected from subsequent sampling in this area. In 1973, the deposition to 5 cm depth at

Sites 25 and 26[10] which are close to Sites A and B was 28,400 and 150,000 mCi/km^2, respectively which is still in reasonable agreement with the values of 45,200 and 103,000 mCi/km^2 obtained from Table 4.

The penetration of plutonium underground illustrated as the percent of the total deposit per cm in Figures 6, 7 and 8, has some striking features. The most striking is that after the maximum is reached, the percent of the total deposit per cm at all sites decreases at a fixed rate per unit depth to about 20-30 cm, whereafter it remains fairly constant. If the decrease is represented as an exponential expression, i e.:

$$A = A_o e^{-\mu d} \qquad (1)$$

then the slopes (μ) at Sites A, B and 7 are 0.33, 0.32 and 0.34, respectively. This trend reinforces the earlier observation of an apparent similarity in the downward transport mechanism at all sites.

The second feature is that at Sites B and 7, the percent of total deposit per cm reaches a maximum at the second sampling depth (between 3.8 and 6.4 cm), while at Site A the maximum occurred within the top 2.5 cm. This indicates that certainly at Sites B and 7 and possibly at A, the plutonium has been transported

downward so that the maximum concentrations no longer exist at the surface. The physical descriptions of the sites suggest that porosity would decrease and plutonium absorbability would increase in going from Sites B to 7 to A which agrees with the penetration patterns in Figures 6, 7 and 8.

The pCi/g of Pu-239-240 at depths greater than 20 cm in Table 3 are at least 30 times greater than the higher blank sample activity reported in Table 2. Therefore, analytical contamination cannot explain the approximately constant activity per gram which we were surprised to observe at each site at the lower depths. While special care was exercised to reduce contamination of the lower depths by surface or near surface debris during collection, contamination as little as 1 part of soil in 1300 could account for the highest activity found below 20 cm at any of the three sites. However, as will be discussed later, the nuclide ratios at the lowest depth samples indicate that contamination by surface debris is not a serious problem. At Site A where the constant percent of total deposit/cm below 20 cm is most apparent, the soil became sticky and plastic at about that depth. Perhaps this is indicative of a change in the soil characteristics which hindered further transport downward and allowed a build-up in the plutonium concentrations.

The Pu-239-240 activity per gram of the composite sticky film samples in Table 5 shows two separate trends. At Site A and D there is some scatter, but the data are fairly constant. At sites B and C there is an increase in the activity per gram with depth. Some of the reduced values for the upper films could be due to the relatively high percentage of dead, non-radioactive vegetation which existed at the surface of the soil to a greater extent at some sites than others.

The following logic can also explain an increase in the activity per gram with depth for some sticky film samples. There is a tendency for large particles in soil to concentrate slightly at the surface. It is likely that the leaking barrels of plutonium contaminated the surfaces of the soil particles, so that the activity per gram of soil would increase with decreasing particle size. There is also a tendency of the sticky film collections to favor the larger particles because of geometric shielding. Consequently, it is not unreasonable for the upper sticky films at some sites to collect an amount of the large particles which is biased slightly high and provide low activity per gram values.

Another factor for the increased activity with depth could be the penetration of the plutonium below ground at a faster rate at some sites. The character of the soil suggests that penetra-

tion would be more likely at Site B than Site A which correlates with the observed trends in the sticky film data and in the data from the soil samples at incremental depths. These data are discussed further in the next section, the Soil Transport Model.

SOIL TRANSPORT MODEL

A semi-empirical model of downward transport of plutonium can describe the observed distributions in the Rocky Flats soil to a depth of about 20 cm. Heat and low humidity can evaporate much of the moisture from the upper levels of the soil. The depth and degree of this evaporative process depends upon the nature of the soil at each site. Subsequent precipitation results in a percolation of water down through this evaporated region permitting a net transfer of plutonium by either solubilization or particle mobilization or both. At lower depths the model assumes that soil moisture approaches a value and consistency, such that further transport of plutonium can be described by a diffusion process through the soil moisture medium to a depth of about 20 cm. Below that, other conditions become important to alter the plutonium transport, and we have not tried to model below 20 cm. However, less than one percent of the total plutonium deposit resides below 20 cm.

The model assumes that the transfer through the top levels of soil where percolation may be taking place follows first order kinetics Consequently, a fixed fraction of the plutonium in any one cm depth interval is exported per year to the next lower cm, while the same fraction of the plutonium in the next upper cm is imported. The magnitude of the fraction and the depth of soil through which this mathematical simulation occurs depends upon the soil characteristics at each site

In the diffusion regime, the percent of the total plutonium deposit per cm follows the equation:[13]

$$\%/cm = 100 \frac{e^{-\frac{Z^2}{4\ Dt}}}{\sqrt{\pi Dt}} \qquad (2)$$

where

Z = depth in cm below the level where the diffusion is assumed to become dominant,

D = diffusion coefficient, and

t = time during which diffusion has occurred.

The airborne concentrations of plutonium just east of the barrel storage area illustrate that the major contamination of land beyond the barrel storage area occurred during clean-up operations in 1968 and early 1969.[1] Consequently for the

samples collected at Sites A, B and 7, in 1975, the time during which the downward transport took place is about 7 years.

The plutonium distributions to a depth of 20 cm were calculated for one-month intervals through a period of 7 years using fractional transfer rates from 10 to 65 percent per cm per year. While some of these calculations described the observed plutonium distributions down to about 6 cm fairly well, they underestimated the observations at the lower levels. By an empirical selection of the depth at which diffusion would apply and a diffusion coefficient of 2 cm^2/year, more reasonable predictions at the lower depths can be made.

The model parameters at each site which give the best fit to the observations are given in Table 7. For example, at Site 7 the plutonium is transported downward from the surface at a rate of 50%/yr per cm to a depth of 6 cm. The amount penetrating 6 cm after the first year is assumed to concentrate at the 7 cm depth from which diffusion via equation (2) is calculated to any depth for a period of 7 years. The amount penetrating 6 cm during the second year is diffused from 7 cm to any depth for a period of 6 years. This procedure is repeated for the remaining 5 years and the estimated values summed for each of the calculated depths.

Figures 6, 7 and 8 illustrate that the model compares reasonably well with the observations. The maximum concentrations are faithfully represented even at Site A in Figure 6 where the first depth sample to 2.5 cm obscures the build-up from the surface to the maximum The diffusion equation is a fair approximation to the transport of plutonium below the maximum concentrations, but tends to give low results at the larger depths. There are slight indications in the observed data that the diffusion coefficient may increase at some depths which would permit more rapid transfer to the lower levels.

The model parameters in Table 7 compare favorably with the soil conditions at each site. Site A appeared to have the fewest stones, the greatest vegetation cover and the highest moisture content near the surface which would tend toward a smaller amount of transfer by the percolation pathway. The best fit parameters in Table 7 for Site A are a fractional transfer rate of 25%/yr cm, and a depth at which diffusion becomes important at 3 cm. Sites 7 and B rate second and third in the above criteria, and the fractional transfer rate increased to 50 and 65%/yr cm, respectively along with an increase to 7 cm for the depth at which diffusion becomes important.

There are two additional sets of observations which tend to verify the model The first is the results of the sticky film

samples. Table 8 gives the integrated pCi/g of the sticky film samples at Sites A and B derived from Table 5 along with the density of the soil in the top profile sample obtained from Table 3. The mCi/km^2 cm can be calculated by dividing the integrated pCi/g by the density and adjusting units. When this value is divided by the integrated mCi/km^2 at the respective site and multiplied by 100, we obtain the percent of the total deposit per cm. Table 8 shows good agreement between the model and the sticky film observations.

The second set of observations which tend to verify the model are the trends in the long term air concentrations at Rocky Flats. One important corollary of this model is that if (at Site 7, e.g.) the plutonium percolates downward through the upper soil levels at 50 percent per cm per year, then the plutonium available for re-suspension at the surface and the attendant airborne concentrations should decrease similarly. From May 1974 to mid 1976, the mean annual air concentrations at Site 4 in Figure 1 decreased with a half life of about 1 year, while at Site 1 from January 1971 to mid 1976 the half life of the annual mean concentration was approximately 2 years.[14] The fractional transfer rates in the upper soil in Table 7 correspond to a surface contamination which would decrease with a half life of 2.4 (25%/yr cm) to 0.66 (65%/yr cm) years in reasonable agreement with the airborne concentrations.

To further test the concepts of this model, we applied it to the measurements of global fallout plutonium with depth at North Eastham, Mass. in 1972 [15] While global fallout has been accumulating on the earth's surface for over a period of 30 years, about half of the cumulative burden in the Northern Hemisphere was deposited by 1962.[16] Therefore, the model calculations were made assuming a single deposit in 1962.

By iteration, good agreement between the observations and calculations as shown in Figure 10 was obtained with a penetration fraction of 15%/yr cm through the top 2 cm and diffusion below 3 cm with a coefficient of 2 cm^2/yr. The divergence from an exponential reduction occurs at about 12 cm at North Eastham which is much closer to the surface than the divergence observed at Rocky Flats sites. This may be real or due to the greater impact of a slight contamination in sampling or analysis of these very low activity level samples However, 93 percent of the total plutonium deposit resides in the top 12 cm at North Eastham.

The soil at North Eastham was a moist, sandy loam with a heavy vegetation cover and virtually no stones or pebbles. It was tilled to 30 cm about 50 years ago, and remained undisturbed since. Owing to the generally lower temperatures and higher humidity at this site as compared to the Colorado sites along with the general character of the soil, it is not surprising that

a lower penetration rate than those observed in Colorado would apply. It is striking that the same diffusion coefficient applies here as it did in Colorado.

This model in its present form is merely a simple mathematical representation of what has already occurred. In fact, the diffusion equation only approximates the behavior of the plutonium from a depth of several centimeters, and the behavior of the very low concentrations below 20 cm is not addressed. Nevertheless, the reasonable results obtained from the application of the diffusion equation with a 2 cm^2/year coefficient to all sites suggests a phenomenon which may be related more to the solubility or transportability of plutonium in soil water than to the characteristics of the soil.

More detailed studies of soil characteristics are required, and additional depth measurements with time at Rocky Flats and other sites are indicated. Such efforts may lead to a better understanding of the long term distribution of transuranics in the environment.

NUCLIDE RATIOS

The Pu-238/Pu-239-240 activity ratio of the soil samples and sticky film samples in Table 6 show no trend with depth under-

ground or site location. The mean value is about 0 018, in good agreement with earlier measurements.[4,10] This result is not surprising since it would be unlikely for isotopes of the same transuranic element in a homogenized source of contamination to behave differently in the environment.

However, the Am-241/Pu-239-240 ratio in the soil samples does increase with depth indicating a slightly greater mobility of the Am-241 in the soil. The maximum enhancement of americium over plutonium is about a factor of 2 at the greatest depths. In 1975, this fractionation effect was slight because over 95% of the total Am-241 at Sites A and B resided at depths where the effect was not apparent. At Site 7, no effect was observed for about 80 percent of the total Am-241 deposit.

The Am-241/Pu-239-240 ratios of the soil samples at the lower depths argue against the probability of serious contamination from the upper levels during collection and sample preparation which had been suggested earlier in the Results section. If such contamination had occurred, the Am-241/Pu-239-240 ratios of the deep samples would reflect the lower ratios of the surface samples rather than the higher ratios observed.

The Am-241/Pu-239-240 activity ratio of the sticky film samples show two separate trends. At Sites A and D there is some

scatter, but the data are fairly constant. At Sites B and C there is a decrease in the ratio with depth. This is the same grouping of sites that was observed in comparing trends in the activity per gram of the sticky film samples in the Results section.

It was pointed out in that discussion and in the section on Soil Transport Model that there was more plutonium penetration at Site B than Site A. Since the Am-241 is slightly more mobile in soil, this effect might partially explain the decrease in the Am-241/Pu-239-240 ratio at Site B. It was also pointed out earlier that there could be differences in the particle size distribution of the sticky film samples with depth. A variation of the Am-241/Pu-239-240 ratio would also have an effect on these results.

The Am-241/Pu-239-240 ratios for the integrated soil columns at Sites A, B, and 7 ranged between 0.11 to 0.13 in good agreement with earlier observations.[10] The ratios for the integrated column for the sticky film samples at Sites C and D agreed with the above value. However, the ratios for the integrated column for the sticky film samples at Sites A and B are about 30% less for reasons which are not clear.

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TABLE 1

EFFICIENCY OF LEACHING AM-241 FROM SOIL

Sample No.	Am-241 pCi per Sample ±% Standard Dev. HASL Value (10^3)	LFE Value (10^3)	LFE/HASL Ratio
P-10	34.1±5	42.4±10	1.24
P-11	12.2±5	13.4±10	1.10
P-12	2.79±3	3.05±10	1.09
P-13	0.653±3	0.757±15	1.16
P-14	0.272±4	0.269±35	0.992
P-15	0.164±2	0.171±50	1.04
P-16	0.0635±5	0.0509±20	0.801
P-28	95.9±6	132±10	1.38
P-29	119±5	136±10	1.14
P-30	58.6±5	67.6±10	1.15
P-31	16.4±5	19.3±10	1.18
P-32	4.36±4	5.05±10	1.16
P-33	3.14±4	3.42±10	1.09
P-34	1.37±4	1.50±10	1.10
P-35	0.168±4	0.192±25	1.15
P-37	0.0604±7	0.0712±35	1.18
P-38	0.242±3	0.246±50	1.02
P-39	0.137±4	0.0878±75	0.639
P-40	0.041±4	0.0347±50	0.856
S-2077*	0.653±3†	0.164±50	0.252‡
S-2078**	4.36±4†	4.91±10	1.12
		Mean	1.08±0.16

*Duplicate of P-13

**Duplicate of P-32

†HASL did not analyze these samples - value of duplicate was assumed.

‡Omitted from mean.

TABLE 2

QUALITY ASSURANCE DATA FOR PLUTONIUM ANALYSIS

Sample No.	Wt. Analyzed (g)	pCi/g ±% Standard Dev. Pu-238	Pu-239-230
		Blank soil samples	
S-2075	132	<0.0002	2.50×10^{-4}±42
S-2076	112	<0.001	4.42×10^{-4}±59
		Duplicate soil samples	
S-2077*	132	0.260±43	10.9±6
S-2077**	"	1.12±43	46.8±6
P-13	118	0.842±7	47.3±3
Mean ±% Standard Dev.†		0.982±20	47.1±1
S-2078	140	5.23±7	266±3
P-32	130	5.45±5	305±2
Mean ±% Standard Dev.		5.36±3	286±9
		Reference soil samples	
S-2079	117		0.0153±5
Expected Value			0.0135
% Deviation			+13
S-2080	116		0.0142±4
Expected Value			0.0135
% Deviation			+5

*Unadjusted for apparent poor leaching efficiency.

**Adjusted for leaching efficiency by multiplying by 1.08/0.252 = 4.29 (see Table 1).

†Mean and standard deviation between results for P-13 and the adjusted S-2077.

TABLE 3

ACTIVITY PER GRAM OF SUB-SURFACE SOIL SAMPLES

Location	Sample No	Depth Range (cm)	Depth Mean (cm)	Area (cm^2)	Weight (g)	pCi/g ±% Standard Dev. Pu-238	Pu-239-240	Am-241
Site A	P-10	0-2.5	1.25	400	500	55.9±9	3000±3	341±5
	P-11	2.5-6.4	4.45		575	15.4±4	833±2	101±5
	P-12	7.0-9.5	8.25		965	2.77±17	191±4	23 4±3
	P-13*	9.5-11.4	10.5		1149	0.982±20	47.1±1	5 54±3
	P-14	12.1-14.0	13.1		1092	0.346±4	18.4±2	2 44±4
	P-15	14.0-16.2	15.1		1343	0.188±15	10.5±3	1 44±2
	P-16	16.2-21.3	18.8	225	1852	0.0896±6	4.91±2	0 581±5
	P-17	21.3-27.9	24.6		2009	0.0230±7	1.21±4	0 18±13
	P-18	28.6-33.3	31.0		1711	0.0198±8	1.13±4	0 17±9
	P-19	33.3-38.1	35.7		1738	0.0175±5	0.905±3	0.21±12
	P-20	38.1-42.5	40.3		1768	0.0174±6	0.968±3	0 15±17
Site B	P-28	0-3.8	1.9	225	180	126±15	7430±3	797±6
	P-29	4.4-6.4	5.4		237	143±13	8290±3	955±5
	P-30	6.4-8.3	7.4		234	80.2±9	4040±4	473±5
	P-31	8.9-10.8	9.8		365	23.2±13	1210±3	131±5
	P-32*	10.8-13.3	12.1		526	5.36±5	286±2	33.5±4
	P-33	13.3-17.8	15.5		923	3.93±6	212±2	25.2±4
	P-34	17.8-23.5	20.6		749	1.73±12	95.9±3	10.2±4
	P-35	23.5-28.6	26.0		1046	0.207±11	10.9±3	1.20±4
	P-36	29.2-34.3	31.7		845	0.0253±9	1.47±5	0.18±10
	P-37	36.2-41.9	39.1		801	0.0622±3	3.47±2	0 455±7

*Mean of duplicate measurements

TABLE 3 (Cont'd)

Location	Sample No.	Depth Range (cm)	Depth Mean (cm)	Area (cm^2)	Weight (g)	pCi/g ±% Standard Dev. Pu-238	Pu-239-240	Am-241
Site 7	P-38	0-3.8	1.9	400	595	0.351±7	18.4±2	2 29±3
	P-39	3.8-5.4	4.6		715	0.160±5	9.05±2	1.09±4
	P-40	5.4-7.0	6.2		714	0.0395±4	2.24±3	0.31±4
	P-41	7.0-8.9	8.0		1157	0.0215±5	1.08±3	0.21±16
	P-42	9.5-10.8	10.1		925	0.0105±10	0.572±5	0.12±19
	P-43	10.8-12.1	11.4		1305	0.00568±6	0.312±2	0.081±24
	P-44	12.1-14.6	13.4		1396	0.00362±10	0.167±3	<0.03
	P-45	14.6-20.3	17.5	225	1846	0.000486±46	0.0406±4	<0.02
	P-46	20.3-26 0	23 2		2056	<0.0004	0.0178±5	<0.02
	P-47	26.4-32.1	29.2		1971	0.000541±33	0.0143±6	<0.02

TABLE 4

AREAL ACTIVITY OF Pu-239-240 WITH DEPTH

Location	Sample No	Depth (cm) Range	Depth (cm) Increment	Depth (cm) Midpoint	mCi/km² cm	mCi/km²	Percent of Total Deposit	Percent of Total Deposit Per cm	Cumulative Percent of Total Deposit
Site A	P-10	0-2 5	2 5	1.25	15,000	37,400	63 7	25 5	63 7
	P-11	2 5-6 4	3 9	4 45	3,080	12,000	20 4	5 23	84 1
	-	6 4-7 0	0 6	6 7	(2,330)	(1,400)	(2 38)	(3 97)	(86 5)
	P-12	7.0-9.5	2 5	8 25	1,860	4,640	7 90	3 16	94 4
	P-13*	9 5-11 4	1 9	10 5	711	1,350	2 30	1 21	96 7
	-	11 4-12.1	0.7	11 8	(450)	(315)	(0 536)	(0 766)	(97 2)
	P-14	12 1-14 0	1 9	13 1	265	504	0 858	0 452	98 1
	P-15	14 0-16 2	2 2	15 1	160	351	0 598	0 272	98 7
	P-16	16 2-21.3	5.1	18 8	79 4	405	0 690	0 135	99 4
	P-17	21 3-27 9	6 6	24 6	16 4	108	0.184	0 0279	99 5
	-	27 9-28 6	0.7	28 3	(17 6)	(12.3)	(0 0209)	(0 0300)	(99 6)
	P-18	28 6-33 3	4 7	31 0	18 2	85 5	0 146	0 0311	99 7
	P-19	33 3-38.1	4 8	35 7	14 5	69 8	0 119	0 0248	99 8
	P-20	38.1-42.5	4 4	40 3	17 3	76 1	0 130	0 0295	100
					Sum	58,700			
Site B	P-28	0-3 8	3.8	1 9	15,600	59,400	23 3	6 14	23 3
	-	3 8-4.4	0 6	4 1	(29,500)	(17,700)	(6 95)	(11 6)	(30 3)
	P-29	4.4-6.4	2 0	5.4	43,700	87,300	34.3	17 1	64 6
	P-30	6.4-8.3	1 9	7 4	22,100	42,000	16 5	8 68	81 1
	-	8 3-8.9	0.6	8 6	(15,500)	(9,300)	(3 65)	(6 09)	(84 7)
	P-31	8 9-10 8	1 9	9 8	10,300	19,600	7 70	4.05	92 4
	P-32*	10.8-13 3	2 5	12 1	2,680	6,690	2 62	1 05	95 0
	P-33	13.3-17.8	4 5	15.5	1,930	8,690	3 41	0 759	98 4
	P-34	17 8-23 5	5.7	20 6	560	3,190	1 25	0 220	99 7
	P-35	23 5-28 6	5.1	26 0	98 8	504	0 198	0 0388	99 9
	-	28.6-29 2	0.6	28 9	(32)	(19.2)	(0 00754)	(0 0126)	(99 9)
	P-36	29.2-34.3	5.1	31 7	10 9	55 4	0.0218	0 00427	99 9
	-	34.3-36.2	1.9	35 3	(15)	(28.5)	(0 0112)	(0 00589)	(99 9)
	P-37	36.2-41 9	5.7	39 1	21 6	123	0 0483	0 00848	100
					Sum	255,000			

*Mean of duplicate measurements

( ) Estimated value from appropriate Figure because no sample was collected.

TABLE 4 (Cont'd)

Location	Sample No.	Depth (cm) Range	Depth (cm) Increment	Depth (cm) Midpoint	mCi / km² cm	mCi / km²	Percent of Total Deposit	Percent of Total Deposit Per cm	Cumulative Percent of Total Deposit
Site 7	P-38	0-3 8	3.8	1 9	72 1	274	49 8	13 1	49 8
	P-39	3.8-5.4	1 6	4.6	101	162	29.5	18 4	79 3
	P-40	5 4-7 0	1.6	6 2	25.0	40 0	7 27	4 54	86 6
	P-41	7.0-8.9	1.9	8.0	16 4	31.1	5.65	2 97	92 2
	-	8 9-9 5	0.6	9 2	(12 2)	(7 32)	(1.33)	(2 22)	(93 6)
	P-42	9.5-10 8	1.3	10 1	10 2	13 2	2 40	1 85	96 0
	P-43	10.8-12.1	1.3	11.4	7 85	10 2	1 85	1 42	97 8
	P-44	12.1-14 6	2 5	13 4	2 34	5 85	1 06	0.424	98 9
	P-45	14.6-20.3	5.7	17 5	0 584	3 33	0 605	0.106	99 5
	P-46	20 3-26 0	5.7	23.2	0 284	1.62	0 295	0.0518	99 8
	-	26.0-26 4	0 4	26 2	(0 246)	(0 0984)	(0 0179)	(0 0448)	(99 8)
	P-47	26 4-32 1	5.7	29 2	0 219	1 25	0 227	0 0398	100
					Sum	550			

( ) Estimated value from appropriate figure because no sample was collected

TABLE 5

ACTIVITY PER GRAM OF COMPOSITE STICKY FILM SAMPLES

Location	Sample No	Sheet Nos.	Sampling Date	Net Wt. (g)	pCi/g Net Wt ±% Standard Dev. Pu-238	Pu-239-240	Am-241
Site A	S-2058	1-3	5/16/75	76.5	36.8±2	2180±2	217±3
	S-2059	4-6		67.0	55.4±2	3330±2	263±3
	S-2060	7-9		67.5	54.1±3	3160±3	254±2
	S-2061	10-12		66.7	47.7±3	2670±3	217±6
	S-2062	13-15		63.5	50.9±3	2860±3	246±3
	S-2063	16-18		61.0	44.9±2	2510±2	220±3
	S-2064	19-21		59.0	42.4±5	2360±5	197±2
	S-2065	22-24		59.4	37.5±6	2110±6	198±2
	S-2066	25-27		33.6	55.0±3	2990±3	324±2
Site B	S-2053	1-3	7/25/73	120.6	18.0±3	1040±3	123±5
	S-2054	4-6		114.4	21.6±5	1200±4	96.4±4
	S-2055	7-9		93.1	27.2±4	1450±4	105±4
	S-2056	10-12		88.6	27.2±5	1430±5	103±4
	S-2057	13-15		82.9	29.1±3	1590±3	114±4
Site C	S-2067	1-3	5/16/73	67.6	0.187±20	10.7±6	1.75±6
	S-2068	4-6		61.8	0.443±18	21.1±7	2.58±5
	S-2069	7-9		58.2	0.491±11	23.7±4	2.65±6
	S-2070	10-12		42.0	0.416±10	20.6±3	2.16±8
	S-2071	13-15		39.4	0.559±10	30.2±4	3.89±5
	S-2072	16-17		38.5	0.905±6	52.3±3	5.00±3
Site D	S-2050	1-3	7/25/73	54.7	1.23±4	59.5±2	8.24±2
	S-2051	4-6		56.9	1.18±4	60.8±2	8.78±2
	S-2052	7-10		75.2	0.829±4	46.4±2	5.36±4

TABLE 6

ACTIVITY RATIOS

Location	Sample No.	Sheet No.	Depth (cm)	Pu-238 / Pu-239-240	Am-241 / Pu-239-240
Site A					
Sticky Films	S-2058	1-3		0.0169	0.0996
	S-2059	4-6		0.0166	0.0789
	S-2060	7-9		0.0171	0.0803
	S-2061	10-12		0.0179	0.0813
	S-2062	13-15		0.0178	0.0858
	S-2063	16-18		0.0179	0.0878
	S-2064	19-21		0.0180	0.0834
	S-2065	22-24		0.0178	0.0938
	S-2066	25-27		0.0184	0.108
	Integrated column			0.0175	0.0872
Soil Samples	P-10		0-2.5	0.0186	0.114
	P-11		2.5-6.4	0.0185	0.121
	P-12		7.0-9.5	0.0145	0.122
	P-13		9.5-11.4	0.0208	0.117
	P-14		12.1-14.0	0.0188	0.132
	P-15		14.0-16.2	0.0180	0.138
	P-16		16.2-21.3	0.0183	0.118
	P-17		21.3-27.9	0.0190	0.15
	P-18		28.6-33.3	0.0176	0.15
	P-19		33.3-38.1	0.0194	0.23
	P-20		38.1-42.5	0.0180	0.15
	Integrated column			0.0183	0.117
Site B					
Sticky Films	S-2053	1-3		0.0174	0.119
	S-2054	4-6		0.0180	0.0805
	S-2055	7-9		0.0187	0.0724
	S-2056	10-12		0.0190	0.0722
	S-2057	13-15		0.0183	0.0717
	Integrated column			0.0183	0.0828

TABLE 6 (Cont'd)

Location	Sample No.	Sheet No.	Depth (cm)	Pu-238 / Pu-239-240	Am-241 / Pu-239-240
Site B					
Soil Samples	P-28		0-3.8	0.0170	0.107
	P-29		4.4-6.4	0.0173	0.115
	P-30		6.4-8.3	0.0198	0.117
	P-31		8.9-10.8	0.0192	0.108
	P-32		10.8-13.3	0.0179	0.110
	P-33		13.3-17.8	0.0185	0.119
	P-34		17.8-23.5	0.0180	0.106
	P-35		23.5-28.6	0.0190	0.110
	P-36		29.2-34.3	0.0172	0.12
	P-37		36.2-41.9	0.0179	0.131
	Integrated column			0.0179	0.113
Site 7					
Soil Samples	P-38		0-3.8	0.0190	0.124
	P-39		3.8-5.4	0.0177	0.121
	P-40		5.4-7.0	0.0177	0.14
	P-41		7.0-8.9	0.0200	0.19
	P-42		9.5-10.8	0.0184	0.20
	P-43		10.8-12.1	0.0182	0.26
	P-44		12.1-14.6	0.0216	<0.2
	P-45		14.6-20.3	0.012	<0.5
	P-46		20.3-26.0	<0.02	<1
	P-47		26.4-32.1	0.038	<1
	Integrated column			0.0186	0.130
Site C					
Sticky Films	S-2067	1-3		0.0175	0.163
	S-2068	4-6		0.0210	0.122
	S-2069	7-9		0.0207	0.112
	S-2070	10-12		0.0202	0.105
	S-2071	13-15		0.0185	0.129
	S-2072	16-17		0.0173	0.0957
	Integrated column			0.0191	0.116

TABLE 6 (Cont'd)

Location	Sample No	Sheet No.	Depth (cm)	Pu-238 / Pu-239-240	Am-241 / Pu-239-240
Site D					
Sticky Films	S-2050	1-3		0.0208	0.139
	S-2051	4-6		0.0194	0.144
	S-2052	7-10		0.0179	0.116
	Integrated column			0.0193	0.133

TABLE 7

MODEL PARAMETERS AT EACH SITE

Site	Fractional Transfer Rate in Upper Soil %/yr cm	Limiting Depth (cm)	Depth at Which Diffusion Begins* (cm)
A	25	2	3
7	50	6	7
B	65	6	7

*Diffusion coefficient is 2 cm^2/yr at all sites.

TABLE 8

COMPARISON OF MODEL WITH STICKY FILM RESULTS

Location	Integrated pCi/g of Sticky Films	Density of Soil in Top Sample (g/cm^3)	$\frac{mCi}{km^2\ cm}$	Percent of Total Deposit Per cm	
				Observed	Model
Site A	2670	0.5	1.3×10^4	23	17
Site B	1310	0.2	2.6×10^3	1.0	0.9

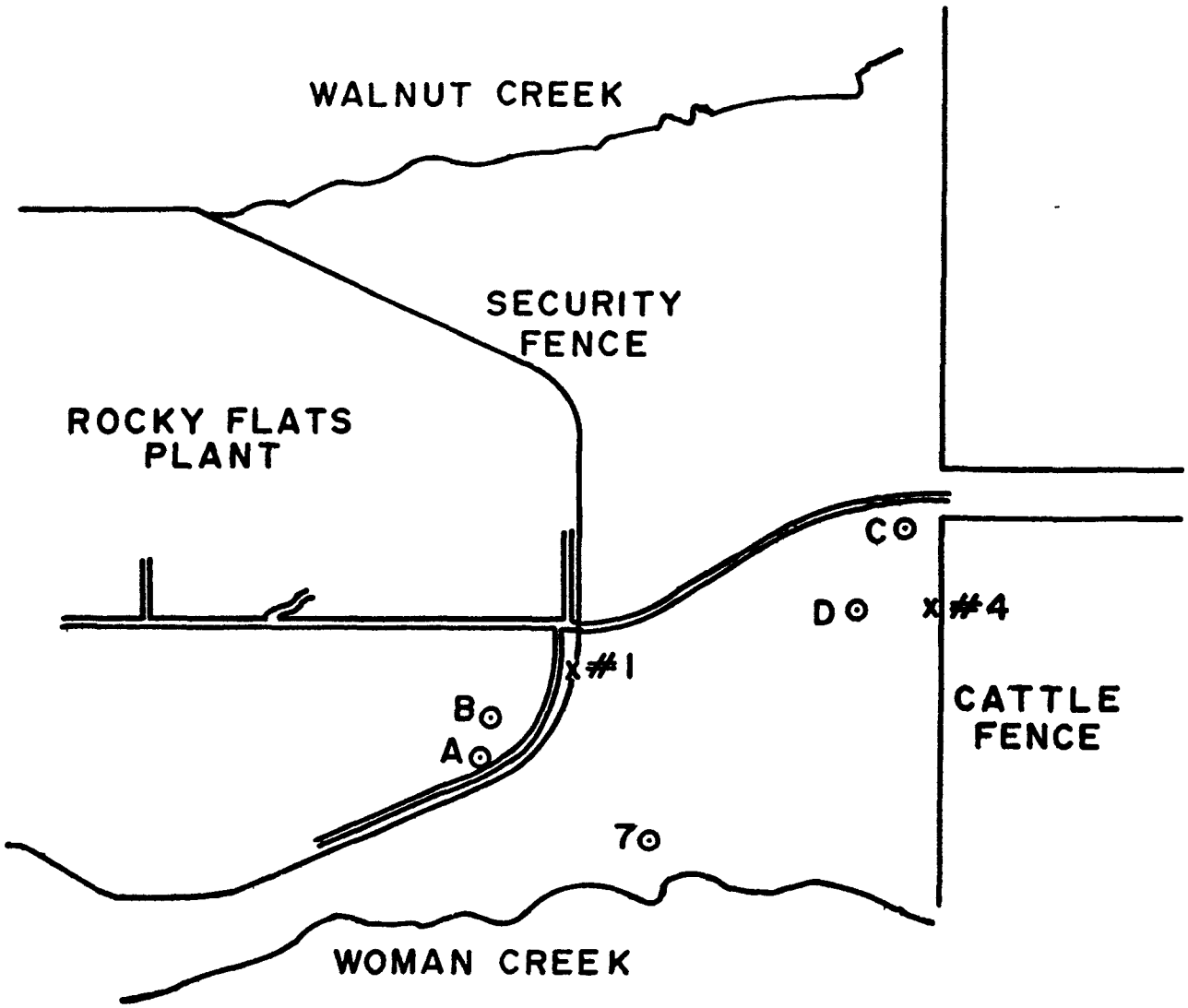

Figure 1. Sampling sites at Rocky Flats.

⊙ Soil sampling
× Aerosol sampling sites

Figure 2. Digging trench in preparation for collecting samples with depth at Site B

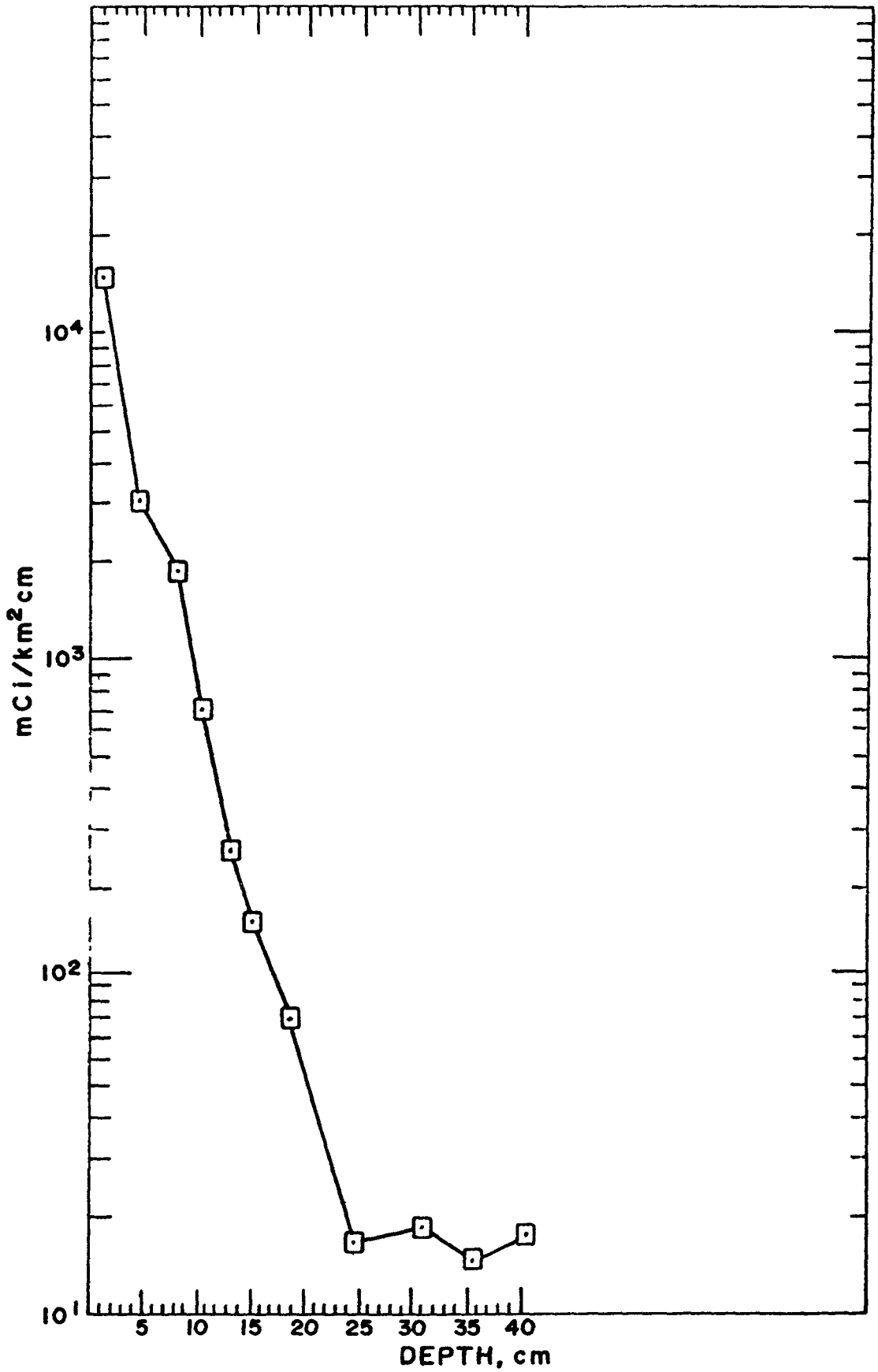

Figure 3. Plutonium deposition per cm with depth at Site A, mCi/km^2 cm.

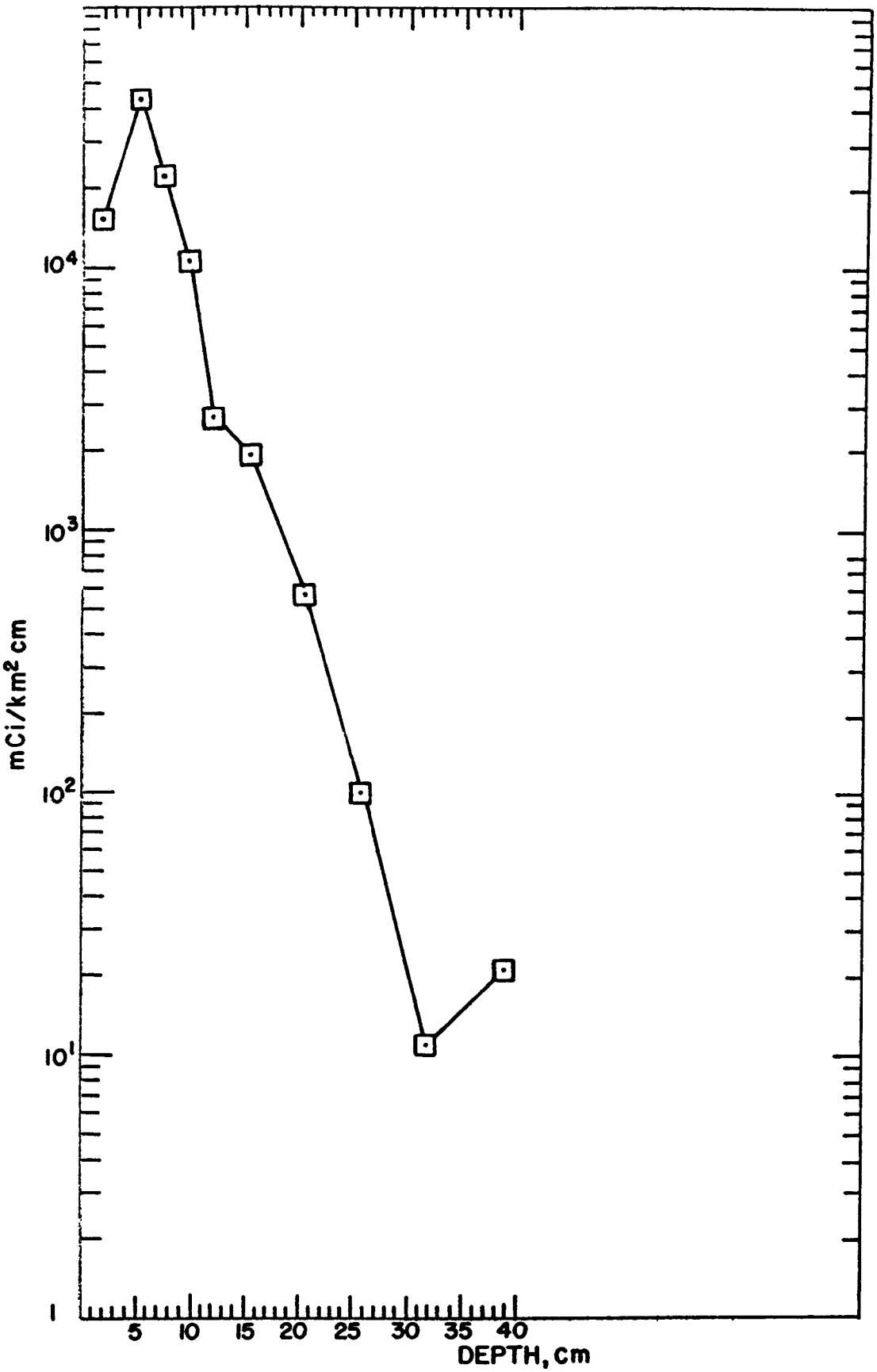

Figure 4. Plutonium deposition per cm with depth at Site B, mCi/km^2 cm.

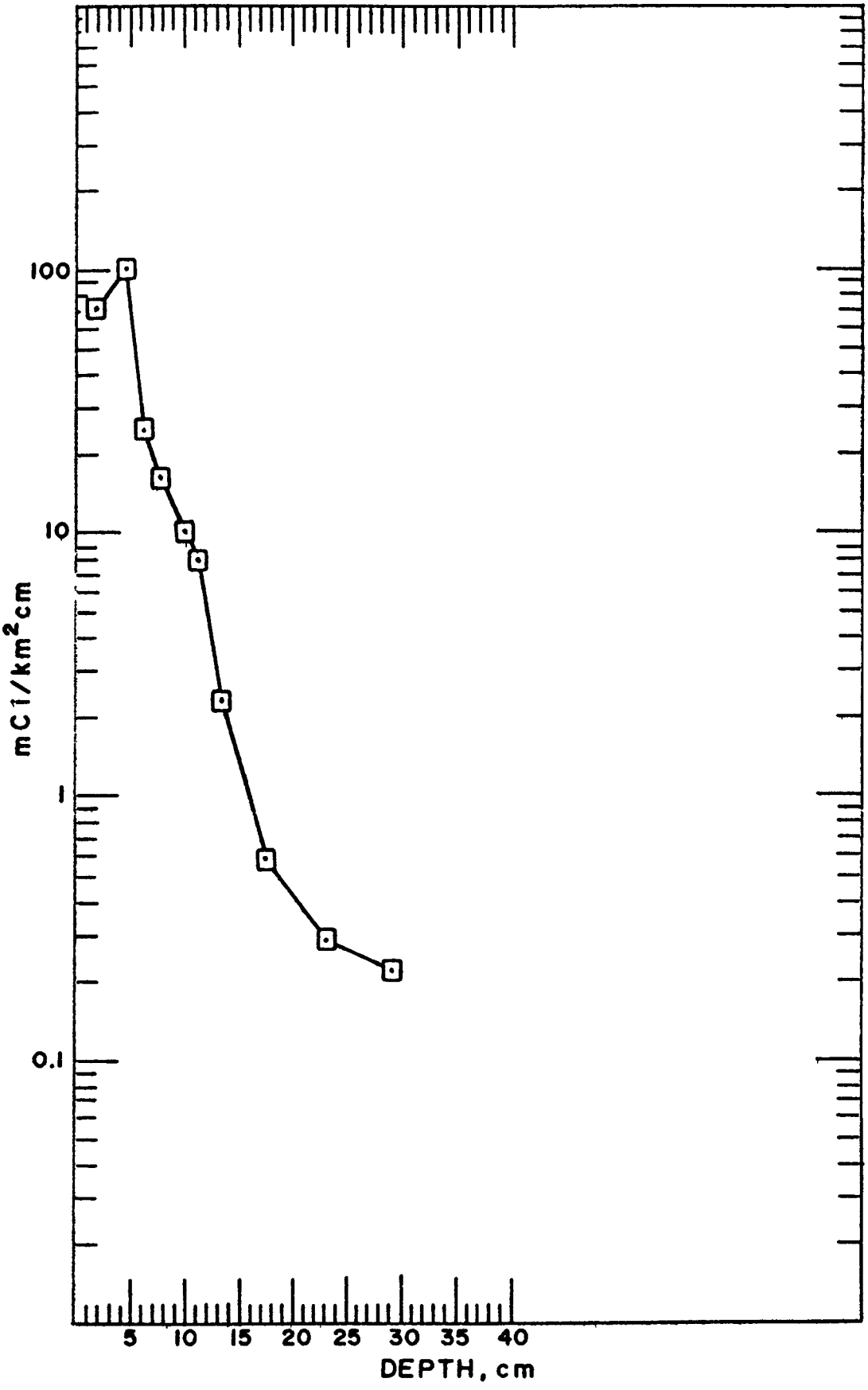

Figure 5 Plutonium deposition per cm with depth at Site 7, mCi/km^2 cm.

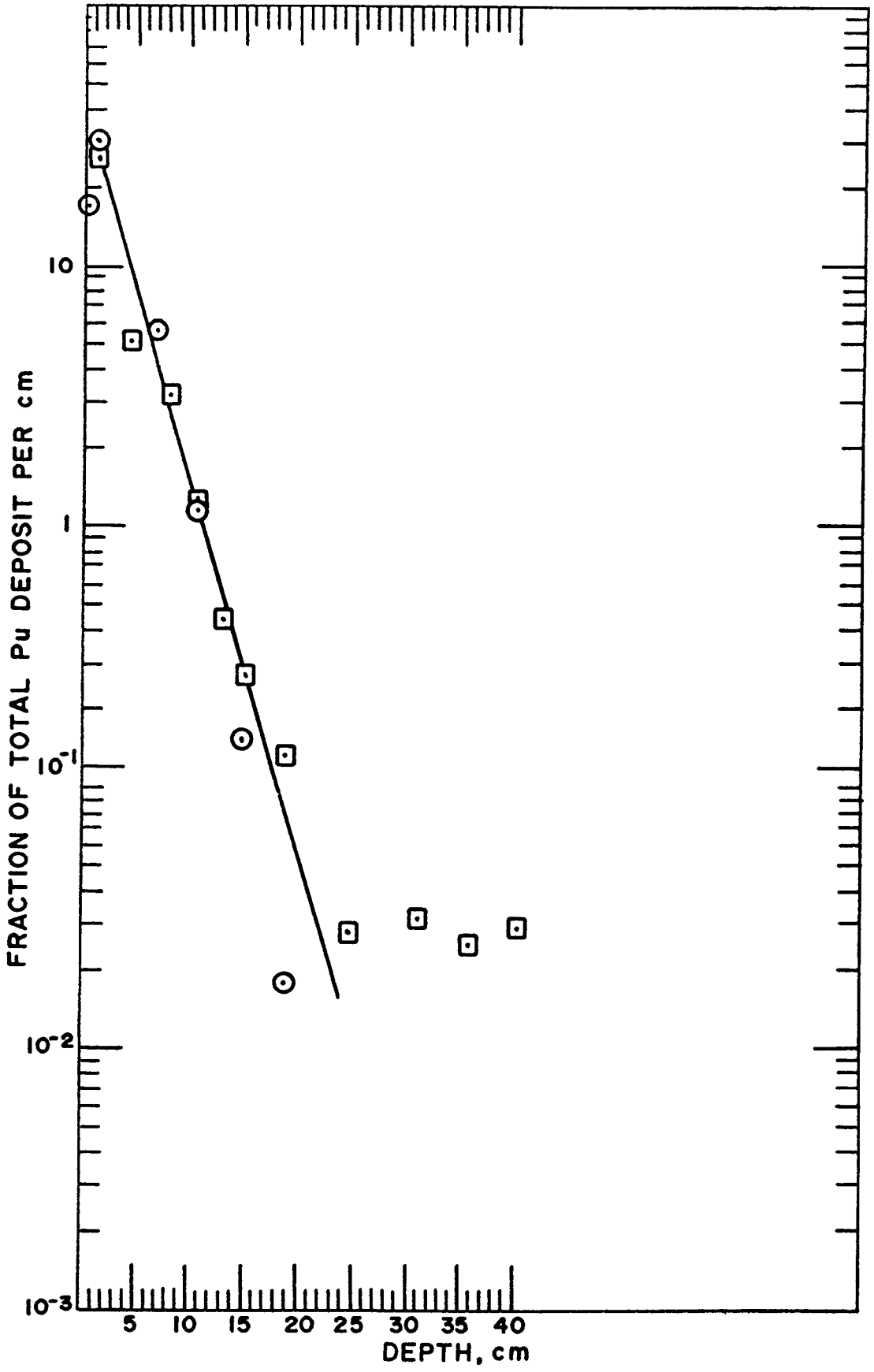

Figure 6. Percent of total plutonium deposit per cm with depth at Site A. - ⊡ - observations; O model calculations using 25%/cm yr penetration through top 2 cm, diffusion below 3 cm with a coefficient of 2 cm^2/yr, and time of 7 years.

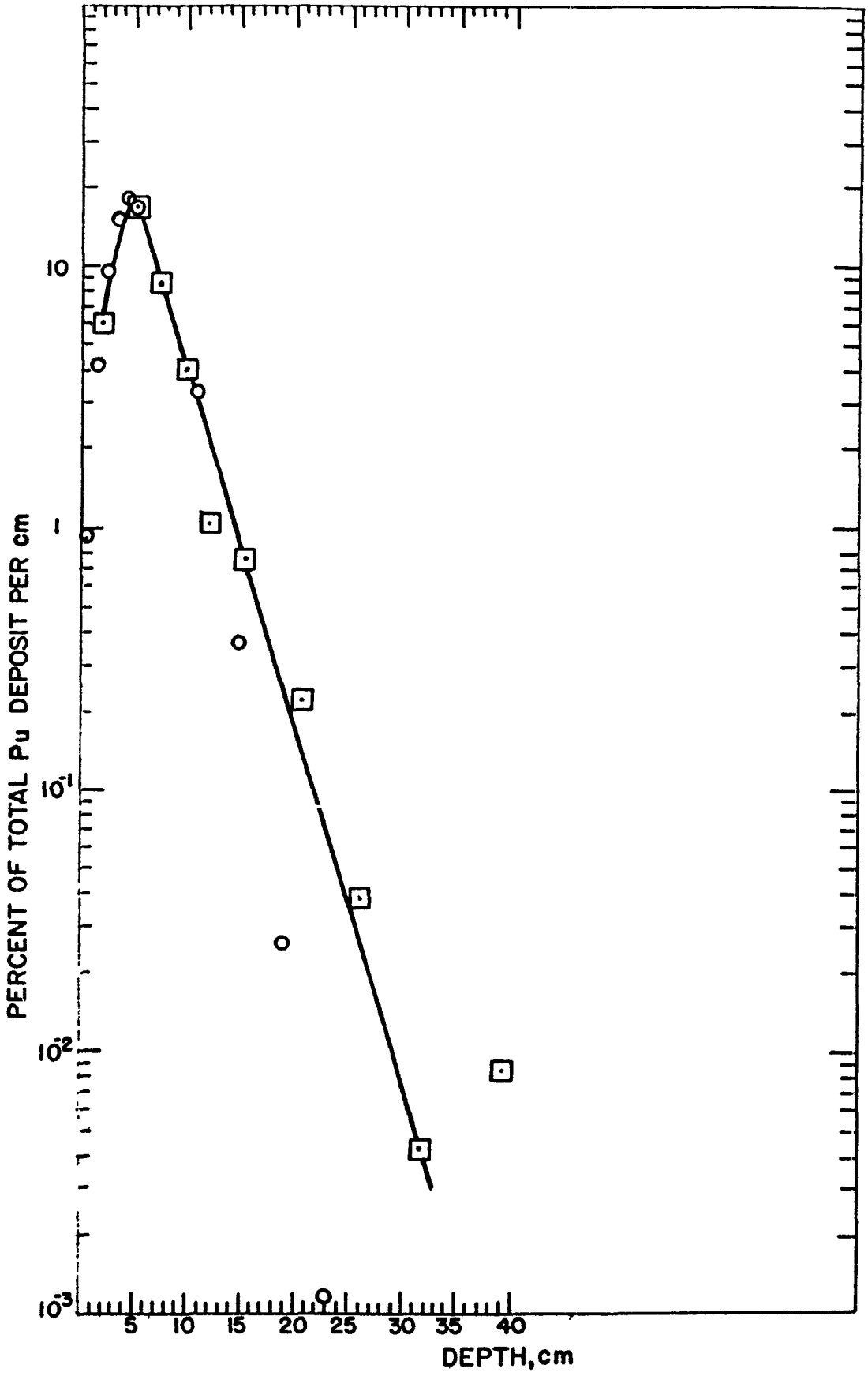

Figure 7. Percent of total plutonium deposit per cm with depth at Site B. - ⊡ - observations; O model calculations using 65%/cm yr penetration through top 6 cm, diffusion below 7 cm with a coefficient of 2 cm^2/yr, and time of 7 years.

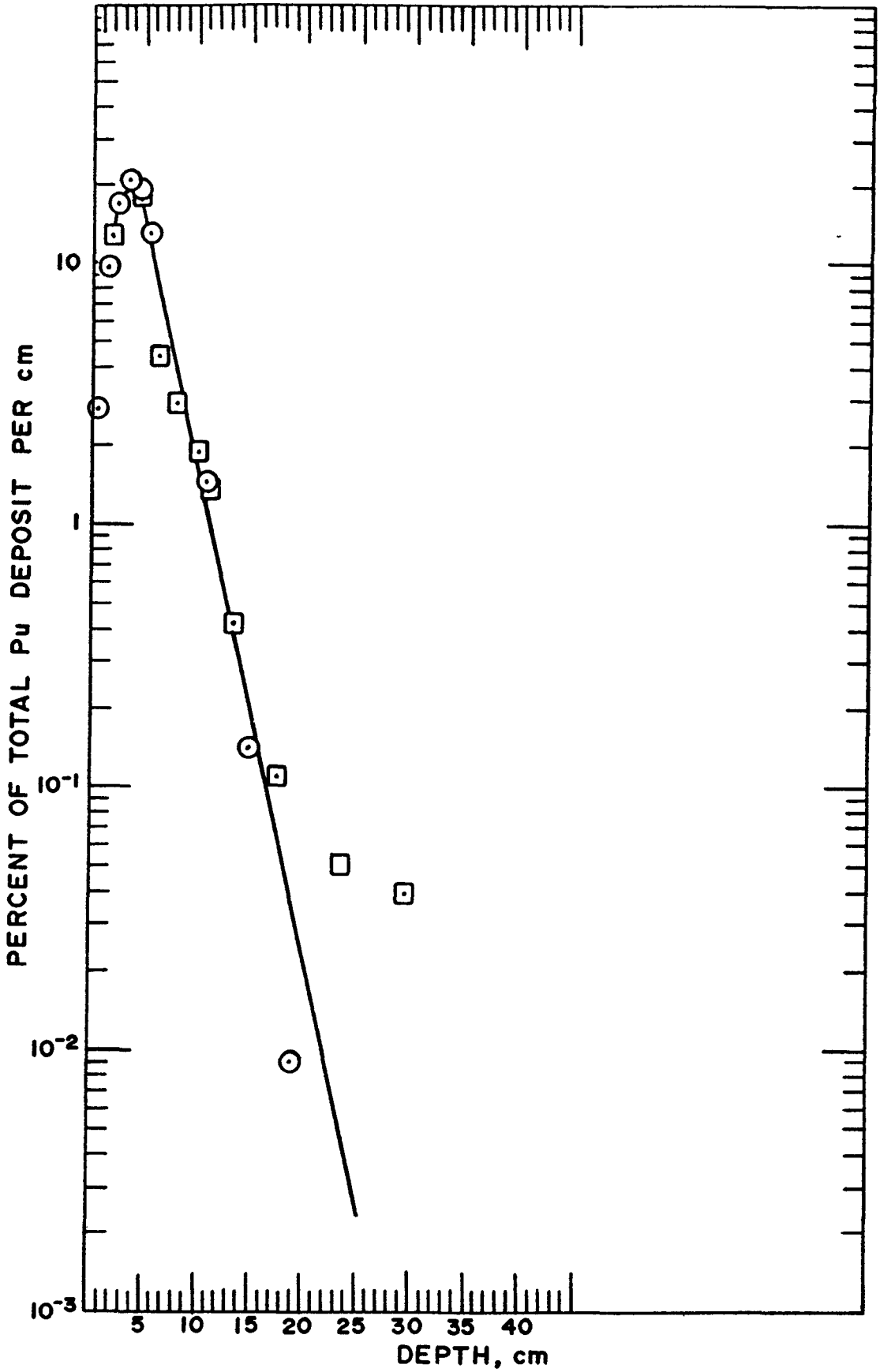

Figure 8. Percent of total plutonium deposit per cm with depth at Site 7. - ⊡ - observations; O model calculations using 50%/cm yr penetration through top 6 cm, diffusion below 7 cm with a coefficient of 2 cm^2/yr, and time of 7 years.

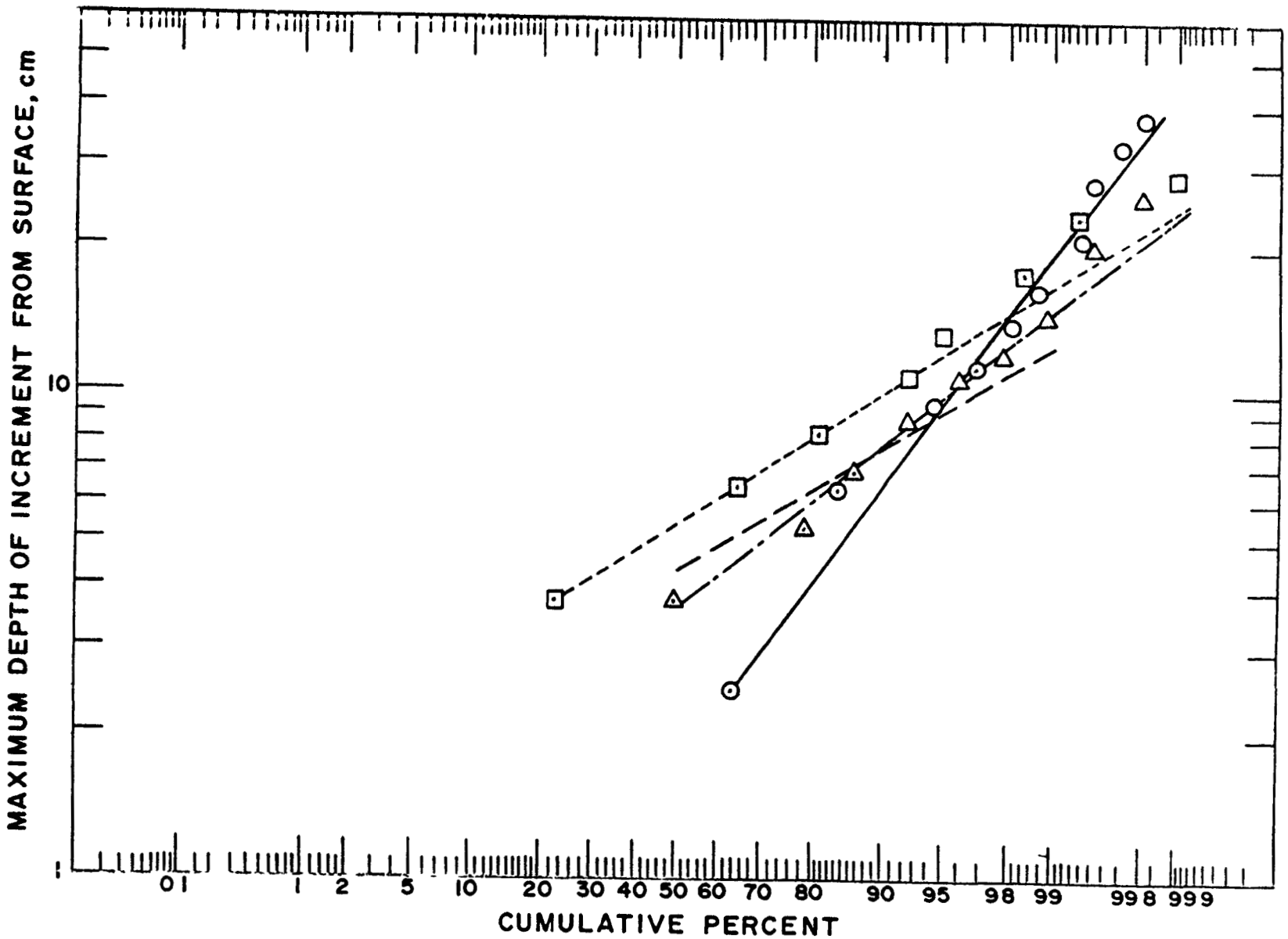

Figure 9 Cumulative percent of total deposit of plutonium with depth; —O— Site A, --□-- Site B, — -Δ- -- Site 7, ---- previous data from HASL-304.

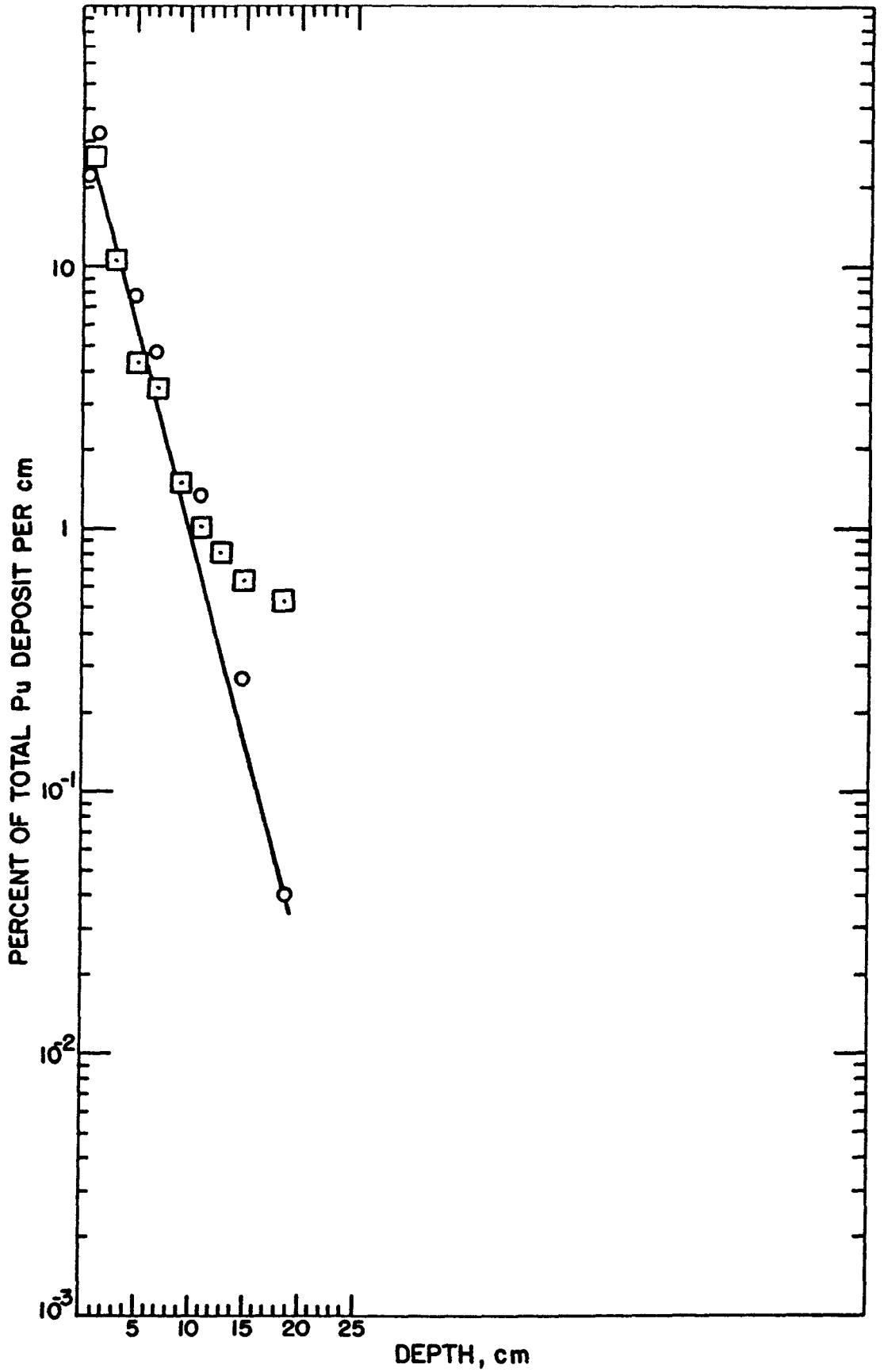

Figure 10. Percent of total plutonium deposit per cm with depth at North Eastham, Mass., 1972. —□— observations, O model calculations using 15%/cm yr penetration through top 2 cm, diffusion below 3 cm with coefficient of 2 cm^3/yr, and time of 10 years.