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QUARTERLY PROGRESS REPORT ON RADIOLOGICAL SAFETY OF PEACEFUL USES OF NUCLEAR EXPLOSIVES: TRANSFER OF TRITIUM TO MAN FROM AN INITIAL WET DEPOSITION

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QUARTERLY PROGRESS REPORT ON RADIOLOGICAL SAFETY OF PEACEFUL USES OF NUCLEAR EXPLOSIVES: TRANSFER OF TRITIUM TO MAN FROM AN INITIAL WET DEPOSITION

M. J. Kelly* and R. S. Booth**

Abstract

The total dose to man from tritium contamination of soil was estimated by simulating the movement of tritium through the soil after an initial wet deposition as a diffusion process with a superimposed translation caused by rainfall. Only experimentally demonstrable constants were incorporated into the model, and it was qualitatively tested against experimental results. Instantaneous equilibrium was assumed between the tritium concentration in the soil compartment and the food that man eats. We concluded that, except for the desert, the time-dependent concentration of tritium in the soil compartment is primarily determined by the rainfall after the deposition, and it is essentially independent of the assumed compartment depth. Consequently, a simple chart could be made for estimating the total dose to man from a given wet deposition source and the subsequent rainfall rate.

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CONTENTS

1. INTRODUCTION

A mathematical model was derived and tested against experimental data to estimate the total dose to man from tritium contamination of soil. This study was made to gain a better understanding of the movement of tritium in the environment. Man needs to understand this phenomenon better because (1) the uptake of tritium, a low-energy beta emitter, is harmful to man; (2) when tritium is released to the environment, it quickly becomes available for uptake by man in the chemical form HTO; and (3), the production of man-made tritium now exceeds that of natural tritium, and this production is directly proportional to the use of nuclear energy, which is expected to increase.

Tritium is primarily an internal radiation hazard to man, because its beta radiation will not penetrate the skin. Because of the abundance of H_2O in the environment, HTO at equilibrium in the hydrological cycle will probably have too small a specific activity to be a hazard to man. Therefore, the greatest hazard of tritium to man will be near a deposition site a short time after the deposition. Since it is a reasonable assumption that the major uptake of tritium will be by food pathways that include the soil, the main objective of this work was to study the movement of tritium through the soil shortly after being transferred to the soil as rainfall (or applied to the surface as a contamination layer).

2. THE MODEL

Most of the tritium released to the environment eventually appears as water vapor. For tritium deposition from rainfall, the vertical integral of the HTO concentration contains about 5 cm^3/cm^2 of water in the tropics, which decreases slowly toward the midlatitudes. In soil having a void volume of 50% (assumed in this study), this initial concentration of 5 cm^3/cm^2 would be distributed over a vertical section 10 cm high. Further distribution would be achieved by translation caused by subsequent rainfall as well as by interdiffusion.

In this study, the average concentration of tritium in the soil was determined over time and space, and then its transfer to man as food was considered. Transpiration and evaporation were not included in the model, but this omission should not alter the results of the calculation significanity.

We solved the space- and time-dependent diffusion equation for the movement of tritium in the soil from an initial concentration in the first 10 cm of soil. The ground surface was the $x = 0$ plane, and the concentration of tritium changed in space only with x, the depth in the ground. The diffusion equation and the

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appropriate boundary conditions are

$$\frac{\partial C(x,t)}{\partial t} = \frac{D\partial^2 C(x,t)}{\partial x^2} \tag{1}$$

$$C(\infty, t) = 0 \tag{2}$$

$$\left.\frac{\partial C(x,t)}{\partial x}\right|_{x=0} = 0 \tag{3}$$

$$\left.\begin{aligned} C(x,t) &= C_0 f/h \quad \text{for } 0 < x \leq h \text{ and } t = 0 \\ C(x,t) &= 0 \quad\quad \text{for } x > h \quad\quad \text{and } t = 0 \end{aligned}\right\} \tag{4}$$

where $C(x,t)$ is the concentration of tritium in Ci/cm^3, D is the diffusion coefficient of water (cm^2/sec), h is the depth of the initial tritium pulse (cm), f is the void fraction of the soil, and C_0 is the initial deposition (Ci/cm^2). The solution[1] to Eq. (1) which also satisfies Eqs. (2), (3), and (4) is

$$C(x,t) = \left(\frac{C_0 f}{2.0h}\right)\left[\operatorname{erf}\left(\frac{h+x}{2\sqrt{Dt}}\right) + \operatorname{erf}\left(\frac{h-x}{2\sqrt{Dt}}\right)\right] . \tag{5}$$

Subsequent rainfall (H_2O) after the initial deposition of tritium exerts a driving force upon this diffusing concentration. This phenomenon was incorporated into our model by superimposing upon this diffusing concentration a vertical translation into the soil. The velocity of this translation, v_g, is related to the rainfall rate r and the void fraction f by the expression $v_g = r/f$. The basis of this relationship is that, as subsequent rainfall fills up the void, the tritium pulse must move farther into the ground.

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[1] W. Jost, Diffusion in Solids, Liquids, Gases, Academic Press, New York, 1952.

Equation (5) with a superimposed vertical translation was programmed on a digital computer, and C(x,t) was evaluated for a sequence of reasonable values for the parameters. After C(x,t) had been calculated at a set of discrete values for x and t, the average concentration of tritium in Ci/cm^3 in the soil from x = 0 to x = root depth, i.e., $\bar{C}(t)$, was determined for each discrete time. The transfer of tritium to man was based on the assumption that instantaneous equilibrium exists between this average concentration in the soil and the food crops, milk, and animal tissue consumed by man.

Equation (1) was also solved with tritium applied to the ground surface as a contamination layer to determine if this difference in the spatial shape of the source would change the results.

3. NUMERICAL VALUES FOR THE CONSTANTS

Only experimentally demonstrable constants were used. A 2000 g/day intake of water from food consumption was used for standard man, and 1 μCi of ^{3}H ingested was considered to deliver a whole body dose of 0.136 mrem. Average rainfall rates were 1 cm/day for the tropics, 0.35 cm/day for the temperate zone, and 0.05 cm/day for the desert. The value of the diffusion coefficient D selected for arable soil with a 50% void fraction (f = 0.5) was 1.0×10^{-5} cm^2/sec. The root depth was 50.0 cm in most calculations.

Figure 1 is a diagram from which D can be calculated for a given soil composition. The diffusion coefficient has a constant value in units of 10^{-6} cm^2/sec along any curve on the diagram, whereas the soil composition is different at each point. The soil composition is determined by the intersection of three percentage composition lines. For example, the point labeled D on the diagram is composed of 60% clay, 20% sand, and 20% silt. The diffusion coefficient for soil of 65% sand, 0% silt, and 35% clay is 1.0×10^{-6} cm^2/sec.

4. RESULTS

A qualitative test of our model against experimental results is presented in Figs. 2, 3, and 4. In Fig. 2 (from Jordan et al.[2]) are plotted observed data that were obtained when tritiated water was added to the surface of a small plot in a tropical rain forest. The theoretical results shown in Figs. 3 and 4 are diffusion plus translation by incoming, uncontaminated rain. In Fig. 3 the deposition was upon

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[2] C. F. Jordan, J. J. Koranda, J. R. Kline, and J. R. Martin, Tritium Movement in a Tropical Ecosystem, UCRL-72146 (December 9, 1969).

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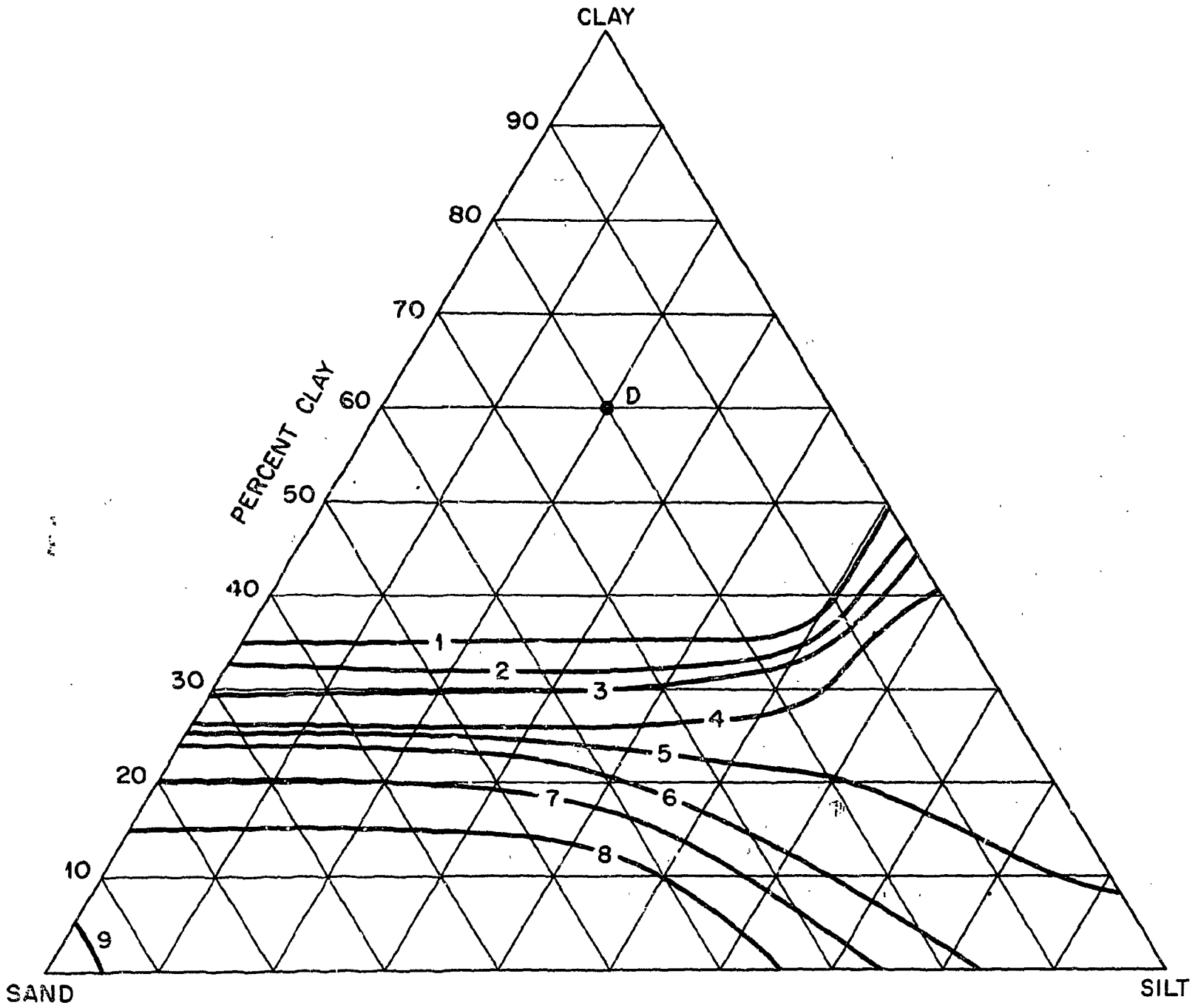

Fig. 1. The Diffusion Coefficient (10^{-6} cm^2/sec) for any Soil Composition.

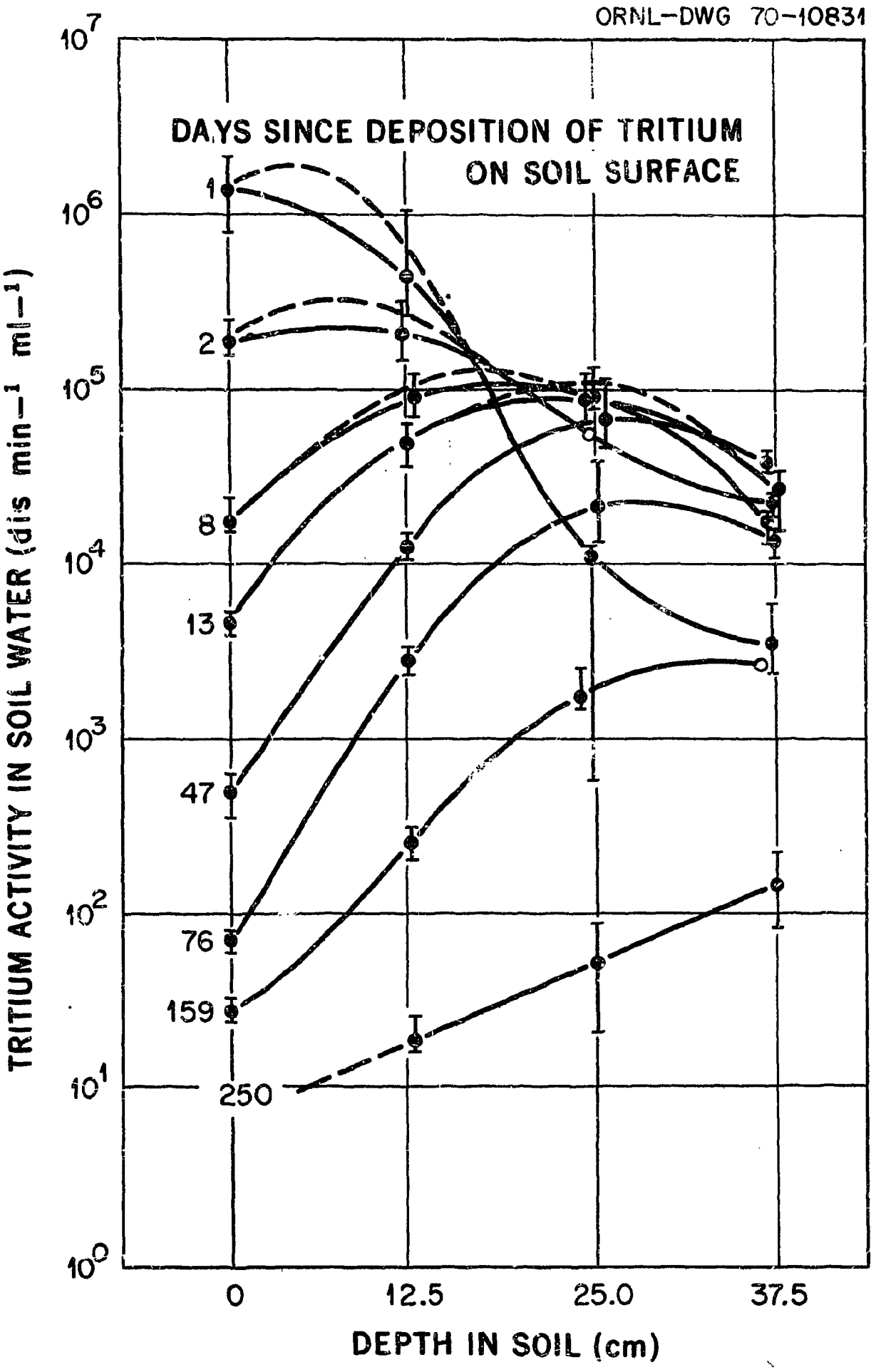

Fig. 2. Tritium Activity in Soil Water as a Function of Soil Depth. (From Jordan et al., UCRL-72146, Fig. 2.)

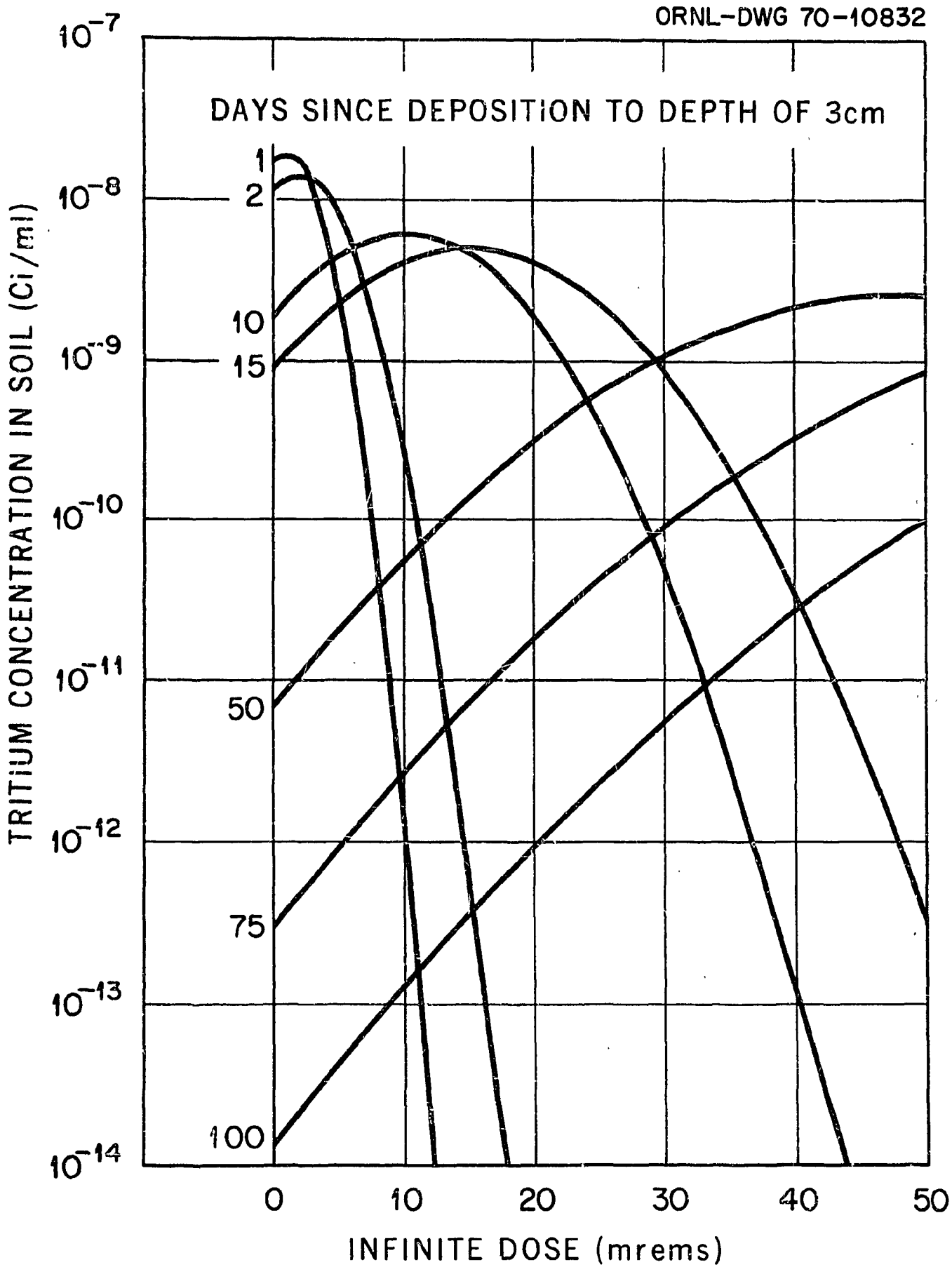

Fig. 3. Tritium Concentration vs Depth in the Soil for Deposition to a Depth of 3 cm.

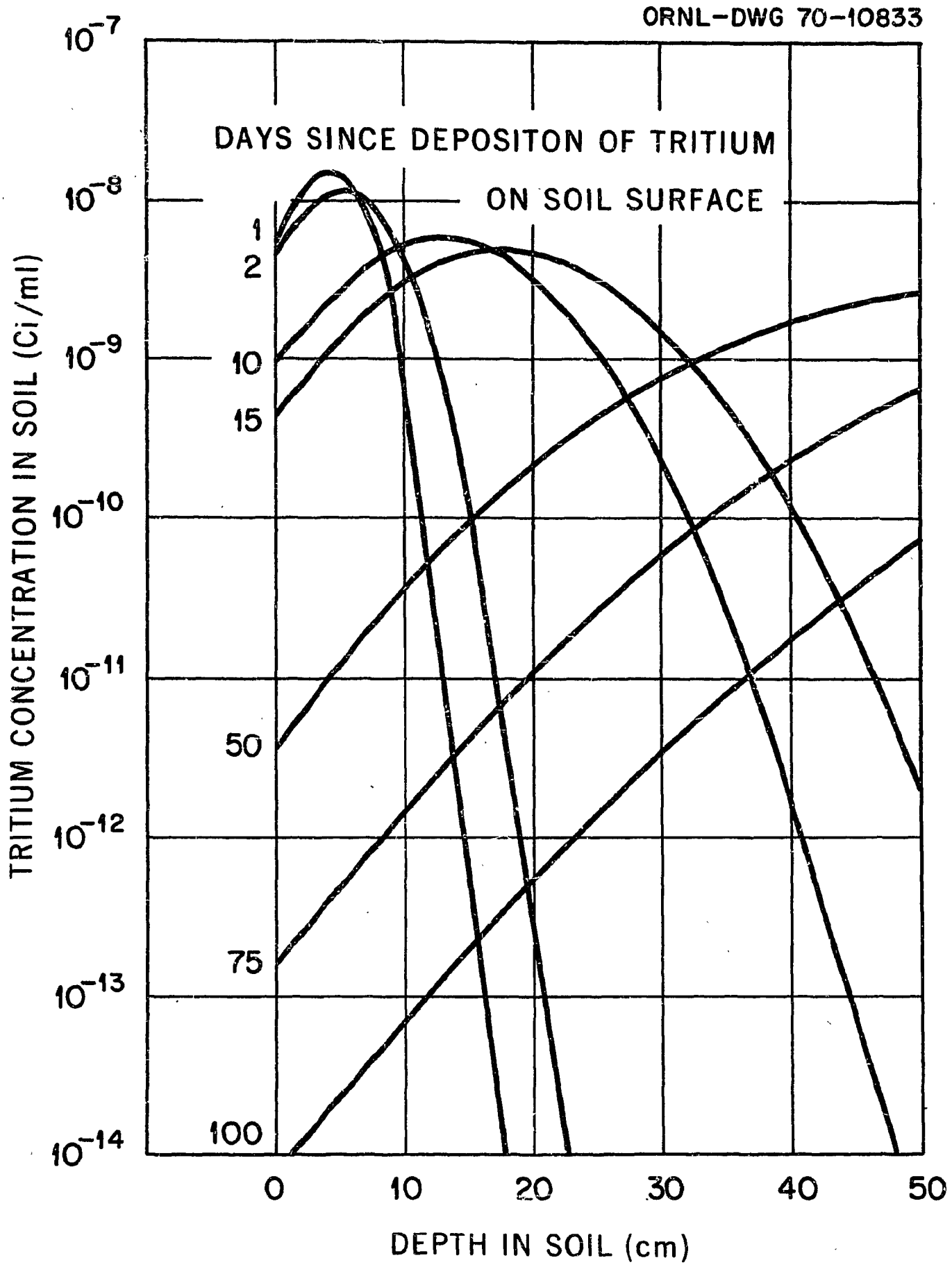

Fig. 4. Tritium Concentration vs Depth in the Soil for Deposition on the Soil Surface.

the soil surface, but in Fig. 4 it was spread out over a range of x from 0 to 3 cm (h = 3). The diffusion coefficient in these calculations was 2.44×10^{-5}, v_g was 1 cm/day, and C_0 was 1×10^{-7} Ci/cm^2.

The real data were an addition to a plot of only 160 by 230 cm located on a slope of about 30°. Thus, lateral drainage not considered in the theory was a significant loss term, and it should cause a more rapid loss from the soil after about the first 2 weeks. Also, a discontinuity in soil composition was encountered at a depth of 25 cm which affected the data beyond 13 days. Therefore, only qualitative comparisons between the theory and experiment are valid.

Jordan et al.[2] state that "For the first 13 days, the peak of the distribution moved downward at a rate apparently equal to the net input of water after application divided by the average water content per unit volume of soil (Fig. 2)." This observation is experimental confirmation of our expression for v_g. The experimentally determined depth of the soil compartment was greater than 38 cm and less than 76 cm, which bound our value of 50 cm. The authors also state that "Although the peak moved through the soil at a rate equal to the calculated transit time, Fig. 2 shows that there was considerable diffusion of tritium away from the peak." This observation confirms the need for incorporation of diffusion as well as translation into the observed data, especially at early times.

The differences in the curves of Figs. 3 and 4 indicate the sensitivity of our results to the space dependence of the initial deposition. As one would expect, the initial source distribution significantly affects our results only when both t and x are on the order of a few days and a few cm, respectively. The qualitative features of the solution are not changed by the initial source distribution.

The importance of the parameters v_g and D is illustrated by the curves in Fig. 5. The concentration of tritium in the soil, C(x,t) Ci/cm^3, is shown as a function of x, with t and v_g as parameters. The constants in these calculations were D = 1.0×10^{-5} cm^2/sec, h = 10 cm, $C_0 = 0.6433 \times 10^{-11}$ Ci/cm^2, and v_g = 0.1 cm/day and 2.0 cm/day. The translation of the peak in the tritium concentration is more clearly shown for the case where v_g = 2.0 cm/day. At t = 10 days, the peak is at 20 cm; and at t = 20 days, the peak is at 40 cm. For v_g = 0.1 cm/day, movement in the peak of the tritium distribution is hardly discernible except for the curve where t = 100 days. Quantitative information about the effect of diffusion can be ascertained for the case with v_g = 0.1 cm/day. The half-width of the tritium pulse at 0.1 its maximum value is 11 cm at 1 day, (20 - 2*) = 18 cm at 20 days, and (41 - 10) = 31 cm at t = 100 days.

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*The value 2 in this expression and 10 in the next were calculated from the expression (v_gt).

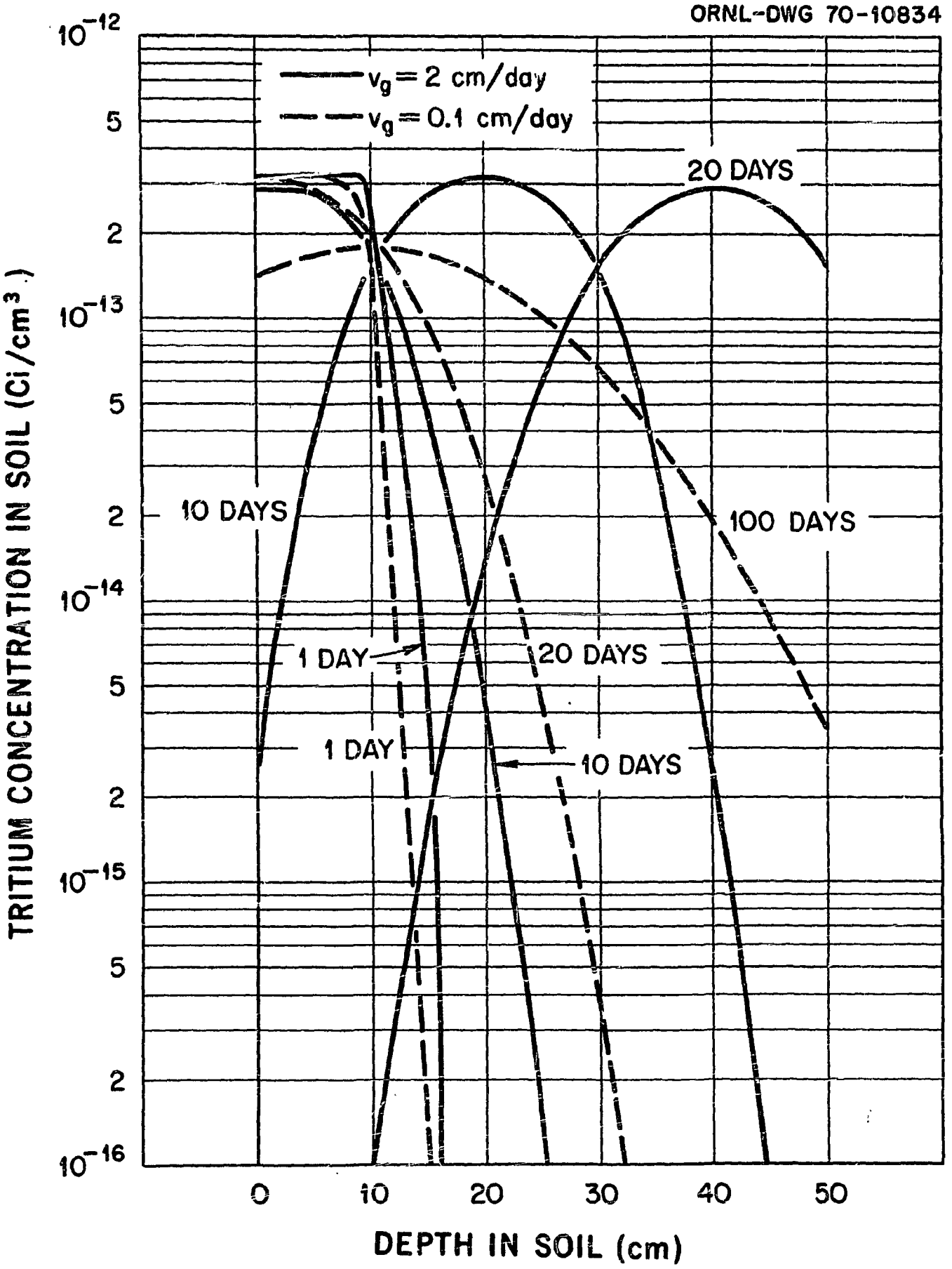

Fig. 5. Tritium Concentration vs Depth in the Soil for Two Values of v_g. The parameter is the number of days since the application of tritium to the soil surface.

A general observation from these curves for a soil compartment depth of about 50 cm and a D of about 10^{-5} is that the loss of tritium from the soil box is primarily by translation out of the compartment for v_g of ~1 cm/day and diffusion is important only for a v_g of less than 0.1 cm/day. This is further illustrated by the subsequent figures and corresponding discussion.

The average concentration of tritium in the soil compartment in Ci/cm^3 was obtained by integrating the tritium concentration from the ground surface to the soil compartment depth and then dividing by this range of integration. This function when multiplied by the intake of water by man in ml/day and divided by the void fraction in the soil yields the time-dependent intake of tritium by man, $\bar{C}(t)$, when the equilibrium assumption of our model is applied. The expression for $\bar{C}(t)$ is

$$\bar{C}(t) = \left(\frac{2000}{0.5}\right)\left(\frac{1}{d}\right)\int_0^d C(x,t)dx \qquad (6)$$

where d is the depth of the soil compartment, and the numerical values for f and man's intake of water have been incorporated into the equation.

Shown in Fig. 6 are curves of $\bar{C}(t)$, with d as a parameter for a rainfall rate typical for a desert. The constants were $D = 1.0 \times 10^{-5}$ cm^2/sec, h = 10 cm, $C_0 = 0.6432 \times 10^{-11}$ Ci/cm^2, and $v_g = 0.1$ cm/day. These calculated results were compared with some of the experimental results presented by Koranda and Martin.[3] They studied the movement of tritium into the soil and its uptake by plants and animals near the SEDAN event which took place in July 1962 when a 100-kiloton thermonuclear device was detonated 635 ft below the surface of the desert in northern Yucca Flat at the Nevada Test Site. Full-scale ecological studies on the persistence of residual tritium at the SEDAN crater were begun in 1966--4 years later. The experimental curves of tritium concentration vs soil depth show the maximum concentration to lie at a depth of between 3 and 4 ft. The shape of these curves closely resembles the shape of the theoretical curve of Fig. 5 for t = 100 days and $v_g = 0.1$. Since all of the tritium profile curves peaked between 3 and 4 ft regardless of ejecta depth, the authors concluded that these distributional patterns were all produced by the same process--rainfall leaching. This conclusion adds further credibility to our model.

Plots of the concentration of tritium in the soil compartment were experimentally determined at several locations by integrating over space the curves of

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[3] J. J. Koranda and J. R. Martin, Persistence of Radionuclides at Sites of Nuclear Detonations, UCRL-71867 (July 15, 1969).

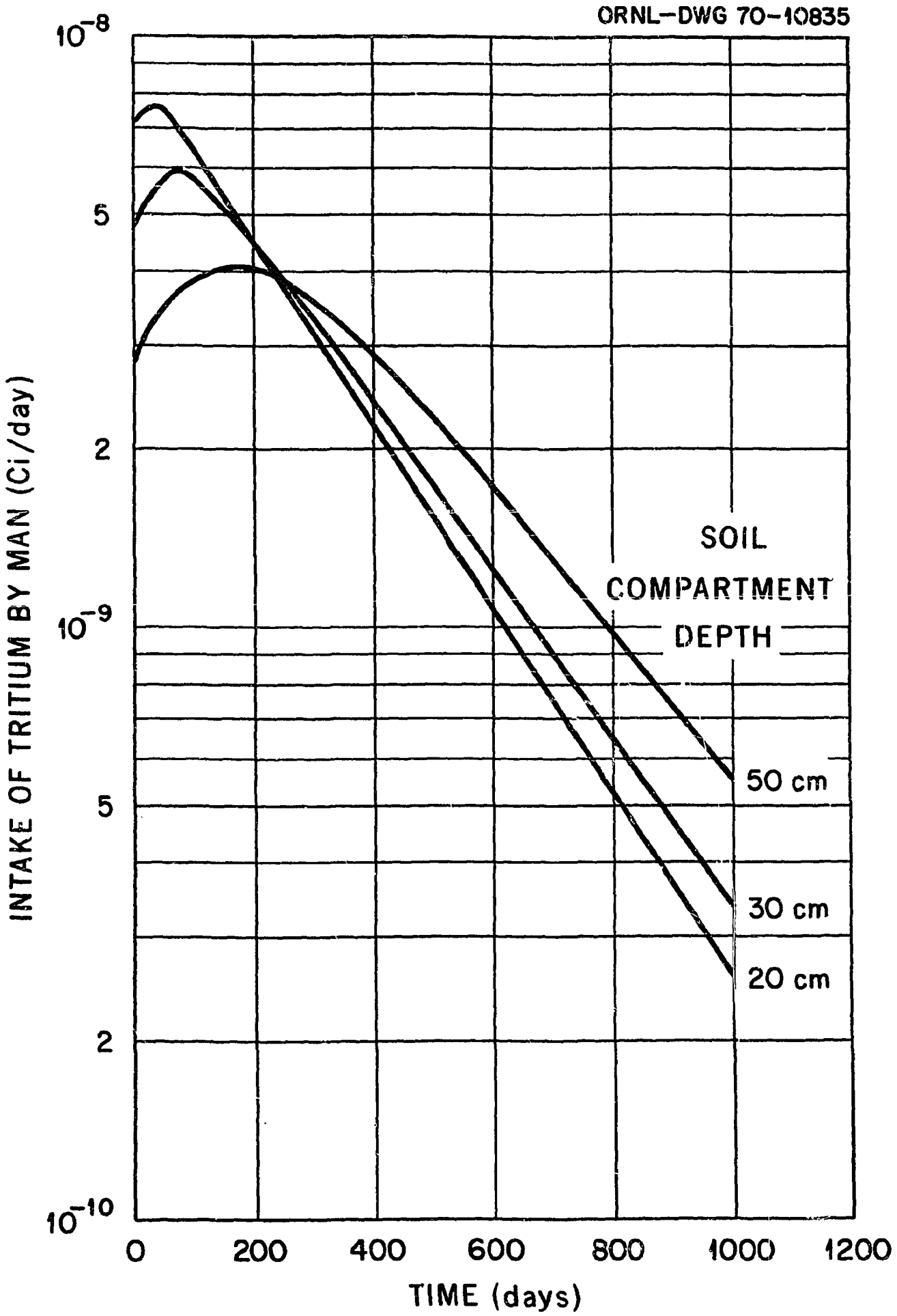

Fig. 6. Intake of Tritium by Man vs Time for $v_g = 0.1$ and for Three Values of Soil Compartment Depth.

tritium concentration vs soil depth. These curves show an exponential decrease with time, and the average for four crater lip sites indicates a mean residence half-time (inverse of the decay constant) for tritium in the SEDAN crater lip of 17 ± 6 months. The variation of these soil-compartment concentrations with compartment depth was not discussed by Koranda and Martin,[3] but it is evident from the curves of concentration vs soil depth that the qualitative behavior predicted by theory in Fig. 6 would have been observed experimentally for d in the range of 3 to 6 ft. It is also consistent with these experimental results that for times beyond about 1 year the soil compartment concentration [$\bar{C}(t)$ times a constant] decreases exponentially with time. Such behavior would be expected when leakage is present in a diffusion process. The half-life obtained theoretically is 7-8 months, which yields a mean residence half-time of 10 to 11.5 months.

The function $\bar{C}(t)$ for $v_g = 0.1$ cm/day is shown in Fig. 7 on a linear scale to t = 200 days. Also shown are plots of $\bar{C}(t)$ for $v_g = 0.70$ cm/sec (temperate zone) and $v_g = 2$ cm/sec (tropics). The dependence of $\bar{C}(t)$ on d (Fig. 6) is not a significant effect for the larger of the two v_g values for d between 10 and 50 cm, and only the curves for d = 50 are shown in Fig. 7. All other parameters are the same in Fig. 7 as they were in Fig. 6. An interesting aspect of the $\bar{C}(t)$ curves for $v_g = 0.7$ and 2.0 cm/day is that, unlike the case for $v_g = 0.1$ cm/day, the loss of tritium from the soil compartment never exhibits an exponential behavior. The loss of tritium is governed more by translation than by diffusion for v_g near 1 cm/day. This is most easily seen for the $v_g = 2.0$ cm/day case, since beyond 25 days, when the peak of the pulse has passed through the soil box, $\bar{C}(t)$ decreases quite rapidly with time.

The cumulative dose to man Q(T) was calculated by multiplying the total input of tritium to time T by the dose conversion factor 0.136 mrem/μCi:

$$Q(T) = (0.136)10^6 \int_0^T \bar{C}(t)dt \qquad (7)$$

where the factor of 10^6 converts Ci to μCi, and the units of Q(T) are mrem. The total integrals of the curves shown in Fig. 7 ($T = \infty$) are 12.2×10^{-9} Ci, 32.2×10^{-9} Ci, and 103×10^{-9} Ci for $v_g = 2.0$, 0.7, and 0.1 cm/day, respectively. Thus, the dose to man is an inverse function of v_g. The integrals of the curves ($v_g = 0.1$ cm/day) shown in Fig. 6 indicate that, for the desert case, the effective annual dose decreases with increasing compartment depth, although the infinite dose is not changed.

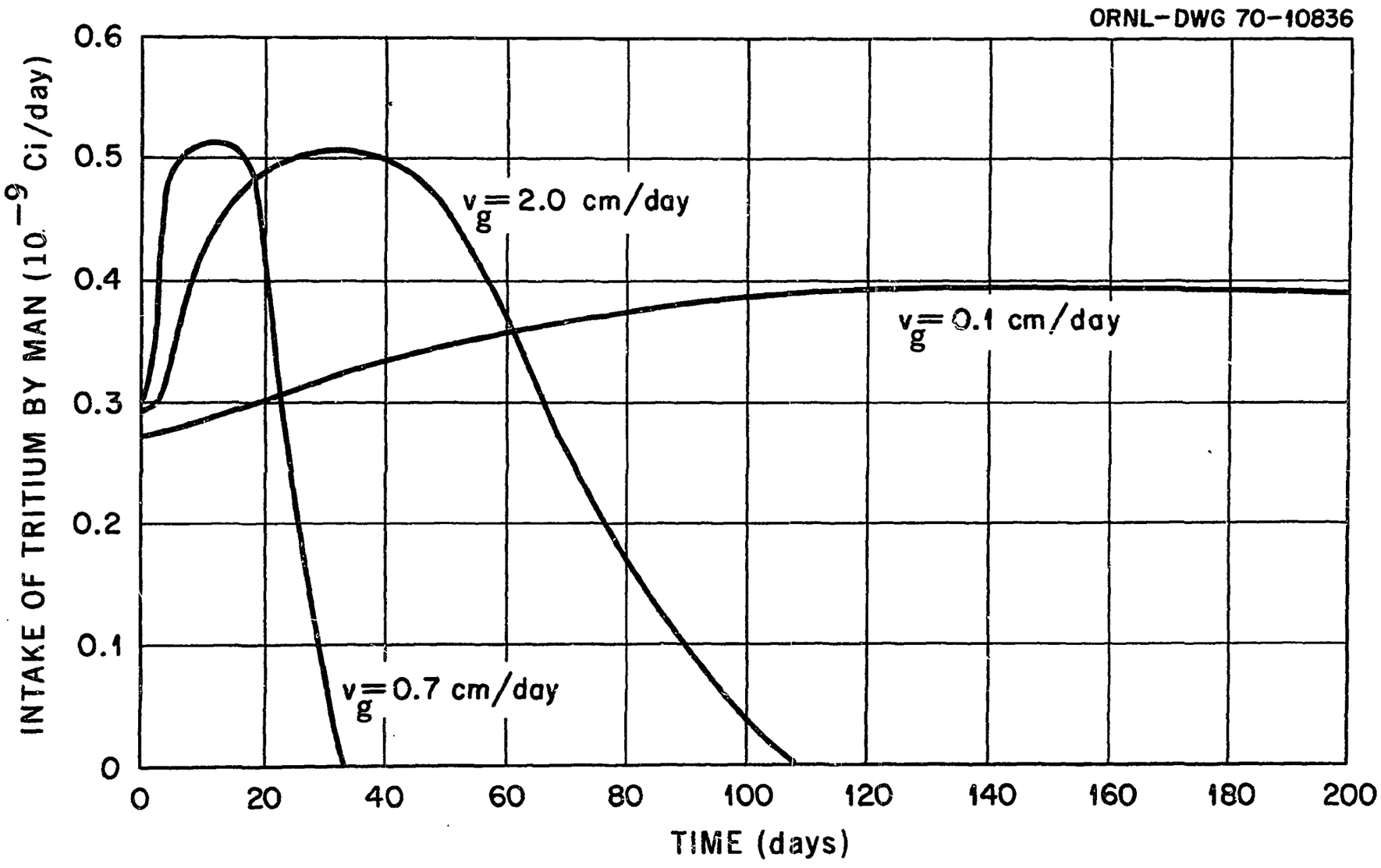

Fig. 7. Intake of Tritium by Man vs Time for v_g = 0.1, 0.7, and 2.0.

In Table 1 values of $Q(\infty)$ for a unit source strength calculated in this study are compared with values calculated by Fleming[4] using the soil-root model developed at LRL.[5]

Table 1. Total Dose to Man, $Q(\infty)$, for a Wet Deposition of 1 pCi/m^2 of ^3H

	Calculated $Q(\infty)$ (rem)	
	by Fleming	by Kelly and Booth
Desert	3.8×10^{-10}	4.4×10^{-10}
Temperate Zone	8.3×10^{-11}	6.3×10^{-11}
Tropics	2.1×10^{-11}	2.2×10^{-11}

These numbers are not completely comparable, because Fleming's values are for a child and a soil-root pathway, and we consider a standard man for all food pathways. However, if our calculations were changed to correspond to Fleming's, they would be higher by less than a factor of 2, a number which is comparable to the accuracy of both models. The importance of the correlation shown in Table 1 is that our results were not achieved by fitting constants and are essentially independent of the soil compartment depth.

The values of $Q(\infty)$ were calculated for this table by numerically integrating $C(x,t)$, [Eq. (5)], over space and time and multiplying the result by constants and conversion factors. The mathematical expressions for this process are given in Eqs. (6) and (7). The $Q(\infty)$ values can be conservatively approximated, with an accuracy of about 25%, in a simple graphical form as shown in Fig. 8. To construct this chart, the calculations were fitted to curves that express a linear relationship between rainfall rate and infinite dose. The justification for such an approximation is as follows: (1) v_g is linearly related to rainfall rate through the expression $v_g = r/f$; (2) for reasonable values of D, losses from the soil compartment as a result of diffusion are significant only for rainfall rates below 0.1 cm/sec; (3) for rainfall rates greater than those of the desert, our results are independent of d, the compartment depth; and (4), if translation is the only means by which tritium can leave the soil compartment, $\bar{C}(t)$ would be constant up to the time when the leading edge of the pulse crossed the bottom of the soil compartment. Beyond this time, $\bar{C}(t)$ would quickly

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[4] Private Communication from E. H. Fleming, LRL, Livermore, California.

[5] Yook C. Ng et al., Prediction of the Maximum Dosage to Man from the Fallout of Nuclear Devices: IV, Handbook for Estimating the Maximum Internal Dose from Radionuclides Released to the Biosphere, UCRL-50163, (May 14, 1968).

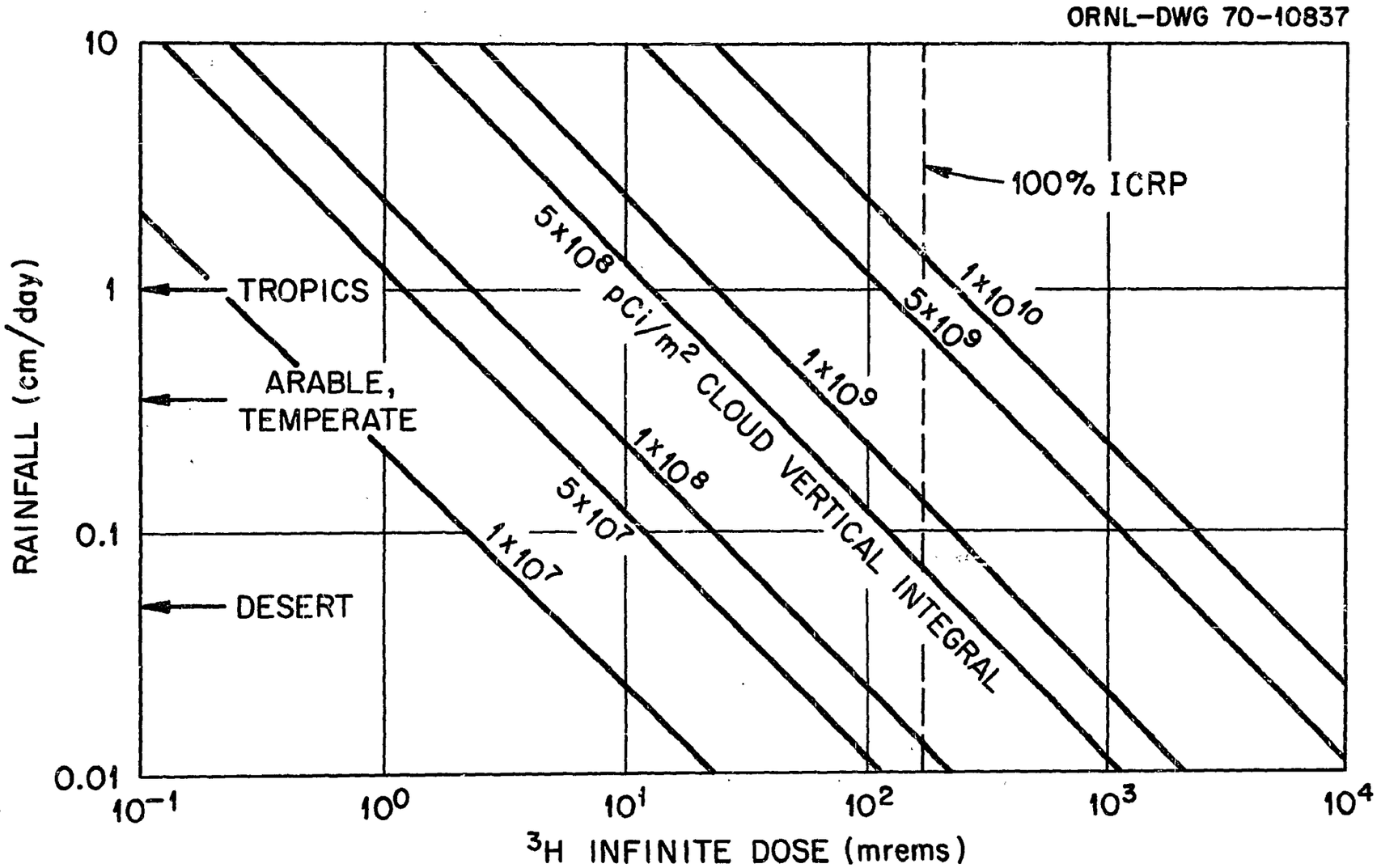

Fig. 8. Chart for Estimating Q(∞) from the Initial Source Strength and the Subsequent Rainfall Rate. Line "100% FRC" is the value of 100% of the dose specified by the Federal Radiation Council.

become zero and remain so. For this case, $Q(\infty)$ would be directly proportional to the transit time of the pulse through the soil compartment. Since the transit time is inversely related to v_g, it is inversely related to the rainfall rate. Therefore, if only translation causes loss of tritium, $Q(\infty)$ is inversely related to the rainfall rate.

In summary, $Q(\infty)$ can be read from a simple chart (Fig. 8). To construct the chart, a functional relationship between rainfall rate and $Q(\infty)$ was assumed. Losses by diffusion were ignored, and thus the chart is increasingly more conservative as the rainfall rate decreases. Within the range of rainfall rates shown on the chart, $Q(\infty)$ values obtained from it will be <25% higher than the value obtained by performing the calculation expressed in Eqs. (6) and (7). The chart applies for the following parameters: $h = 10$ cm, $D = 1 \times 10^{-5}$ cm^2/sec, $f = 0.5$, and an input to man of 2000 g/day of water from the food he eats. For convenience in interpolation, the correlation between rainfall rate and $Q(\infty)$ is shown for several values of the initial wet-deposition source, which is expressed in units of pCi/m^2.

5. ACCURACY OF THE MODEL

The accuracy expected from this model depends on the particular environmental condition against which it is tested. We believe the model to be conservative. The only nonconservative assumption knowingly made was to consider tritium transferred to man only by the food he eats. Contamination of his water supply was ignored, since the specific concentration of tritium in the water supply would normally be much lower than in the food produced on land exposed to the deposition. This assumption causes less than a 50% error.

Assumptions which tend to make the model conservative include the neglect of evaporation and runoff. The total error caused by these omissions depends a great deal on the environmental situation, but it should be less than a factor of 2. Also, we assumed instantaneous equilibrium between man's food supply and the soil compartment. The assumption introduces comparatively little error for food crops that are continuously harvested and for milk, but the model could be quite conservative for meat and for food crops that are only occasionally harvested. These results could, in a given situation, easily be 50% too low or a factor of 10 too high.

6. CONCLUSIONS

We developed a model for predicting the movement of tritium to man from an initial wet deposition and gained insight into the parameters that are most important

in this transfer. The model agrees qualitatively with existing experimental results and requires only experimentally demonstrable constants. A more detailed and more accurate model could be derived for a particular environmental situation where meterological and hydrological data are available and if accurate transfer rates of tritium were available for the crops and animals that constitute man's diet.