

# **Migration of radionuclides in fissured rock:**

**Some calculated results obtained from a model based on the concept of stratified flow and matrix diffusion**

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MIGRATION OF RADIONUCLIDES IN FISSURED ROCK:  
SOME CALCULATED RESULTS OBTAINED FROM A MODEL  
BASED ON THE CONCEPT OF STRATIFIED FLOW AND  
MATRIX DIFFUSION

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This report concerns a study which was conducted for SKBF/KBS. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client. -

A list of other reports published in this series during 1982, is attached at the end of this report. Information on KBS technical reports from 1977-1978 (TR 121), 1979 (TR 79-28), 1980 (TR 80-26) and 1981 (TR 81-17) is available through SKBF/KBS.

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## SUMMARY

Some computed results of radionuclide migration in fissured rock are presented. The computations are based on a model which describes flow as occurring in a multitude of independent fissures (stratified flow). This gives rise to strong "dispersion" or channeling. The radionuclide migration in the individual fissures is modelled by the advection equation in a parallel walled channel with porous walls. The nuclides may diffuse into the pores and sorb reversibly on the pore surfaces.

The effluent rates of 23 important nuclides are presented as functions of distance and time for various values of important parameters such as rock permeability, diffusion coefficients, release rates, time of first release, fissure spacing and fissure width distribution.

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## 1. BACKGROUND

Radionuclides which escape from a final repository for high level waste in deep geologic media, will have to migrate through the bedrock to reach the biosphere. In the Swedish concept (KBS 1978) the repository will be in crystalline rock at 500 m depth. The bedrock has a low hydraulic conductivity. Flow of water occurs in fissures in the rock. The radionuclides may interact in several ways with the bedrock. The rock has a low connected porosity of 0.1 - 1 % (Brace 1965, Skagius 1981). The radionuclides may migrate into these pores by diffusion (Neretnieks 1980, Skagius 1981) and may sorb on the pore surfaces. Nonsorbing species may utilize the micropore volume for retardation also, as this pore volume will give an additional residence time, where the radionuclide will have more time to decay.

The water conducting fissures in the bedrock may have fissure coating material e.g. chlorite, calcite (Larson et al. 1981) and fissure filling material, e.g. montmorillonite clay. These substances may also add to the retardation of the sorbing nuclides.

In an individual fissure a nuclide will thus be retarded in relation to the flow velocity of the water by the following mechanisms:

- o Sorption on fissure filling and coating material.  
This is summarily called surface sorption and is quantified by a surface sorption coefficient  $K_a$  [m].  
 $K_a$  indicates the sorption capacity of a nuclide per surface area of fissure.
- o Migration into the micropores of the rock by diffusion.  
The diffusivity in the micropores  $D_p$  determines the rate of migration into the pore volume, and the porosity  $\epsilon_p$  determines the volume which may be accessed.
- o Sorption on the inner surfaces of the rock (microfissure surfaces). This is summarily called volume sorption and is quantified by a volume sorption coefficient  $K_{d,p}$  m<sup>3</sup>/m<sup>3</sup>.

As a first approximation and due to the lack of better data for most radionuclides, the sorption coefficients  $K_a$ ,  $K_d \rho_p$  and the diffusivity  $D_p$  for every nuclide is assumed to be constant, independent of concentration and time.

Different fissures may have different flow velocities, and the set of individual pathways from the repository to the biosphere which are made up of individual connected fissures, may also have considerable variations in travel times. In porous media such variations are described by the term hydrodynamic dispersion. In sparsely fissured rock individual channels may be poorly connected to others. In extreme cases the bedrock might be seen as consisting of a series of independent channels which all have different water velocities and flowrates. Such cases exhibit a "dispersion" behaviour entirely different from "normal" hydrodynamic dispersion as the fast channels do not dilute their nuclides in the water of the slower channels. If such channeling occurs, the fast channels may carry the nuclides much faster than the average water velocity of the bedrock. The time for decay might in some cases be so much less as to make a significant difference in the effluent concentrations. (Neretnieks 1981)

The model used in this study includes the effects of channeling (or stratified flow), surface sorption, matrix diffusion and volume sorption.

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## 2. MODEL USED FOR COMPUTATIONS

The derivation of the model is given in extenso in (Neretnieks 1980 and Neretnieks et al. 1981). A summary is given below.

### Individual fissure

In an individual fissure of width  $\delta$  where the fissure walls are porous, the tracer concentration  $c_f$  in the fluid in the fissure is determined by

$$\frac{\partial c_f}{\partial t} \cdot R + U_f \frac{\partial c_f}{\partial x} = \frac{2D_e}{\delta} \cdot \frac{\partial c_p}{\partial z} \Big|_{z=0} \quad (1)$$

$$R = 1 + \frac{2K_a}{\delta} \quad (2)$$

$R$  is the retardation factor due to surface sorption.

$U_f$  is the water velocity in the fissure.

$D_e$  is the effective diffusivity in the micropores.

$z$  is the depth into the wall.

$x$  is the distance along the fissure.

The last term describes the loss of tracer from the fluid flowing in the fissure due to diffusion into the porous matrix of the wall. In this model hydrodynamic dispersion is neglected. The diffusion into the matrix is given by

$$\frac{\partial c_p}{\partial t} = D_a \frac{\partial^2 c_p}{\partial z^2} \quad (3)$$

where

$$D_a = D_e / K_d \rho_p$$

and  $c_p$  is the concentration in the fluid in the pores.

The initial and boundary conditions are: Decaying step input at the inlet ( $x=0$ ) starting at time  $t_0$  and ending at time  $t_0 + \Delta t$ . The bedrock is initially free of nuclide. This can be written

Initial condition.

$$c_p = c_f = 0 \quad t < t_0 \quad \text{all } x \text{ and } z \quad (4)$$

Boundary condition 1

$$c_p = c_f = 0 \quad \text{when } t > 0 \quad \text{for } z \rightarrow \infty \quad (5)$$

Boundary condition 2

$$c_f = c_0 e^{-\lambda t} \quad \text{for } t_0 < t < t_0 + \Delta t \quad \text{at } x = 0 \quad (6)$$

$$c_f = 0 \quad \text{for } t > \Delta t \quad \text{at } x = 0$$

The solution to these equations have been obtained by the Laplace transform method (Neretnieks 1980). The solution is

$$c_p / c_0 = e^{-\lambda t} \operatorname{erfc} \left[ \frac{G}{(t - t_0 - Rt_w)^{1/2}} \right] - \operatorname{erfc} \left[ \frac{G}{(t - t_0 - \Delta t - Rt_w)^{1/2}} \right] \quad (7)$$

where

$$G = \left\{ \left[ D_e + \frac{1}{2} \frac{U_f \delta z}{x} \right] / \delta D_a \right\}^{1/2} t_w \quad (8)$$

The solution applies for  $z > 0$  and  $x > 0$  and for  $t > Rt_w + t_0$  for the first erfc expression and for  $t > Rt_w + t_0 + \Delta t$  for the second erfc expression. Otherwise these expressions are 0.



The concentration in the fissure  $c_f$  is obtained for  $z = 0$  and becomes

$$c_f/c_o = e^{-\lambda t} \left\{ \operatorname{erfc} \left[ \frac{1}{H^{1/2} (t - t_o - Rt_w)^{1/2}} \right] - \operatorname{erfc} \left[ \frac{1}{H^{1/2} (t - t_o - \Delta t - Rt_w)^{1/2}} \right] \right\} \quad (9)$$

where

$$H = \frac{\delta^2}{t_w^2 D_e K_d \rho_p} \quad (10)$$

as

$$D_a = D_e / K_d \rho_p \quad (11)$$

The entities  $\delta$  and  $t_w$  in equation 10 are difficult to measure in the field. They may be determined from other entities which are more amenable to observations.

With

$$t_w = x/U_f, \quad U_f = U_o/\epsilon_f$$

and

$$\epsilon_f = \delta/S \quad \text{where } U_o \text{ is the water flux } m^3/m^2 \text{ of bedrock} \cdot s$$

$S$  is fissure spacing

we obtain

$$\frac{\delta}{t_w} = \frac{\delta U_f}{x} = \frac{\delta U_o}{\epsilon_f x} = \frac{S U_o}{x} \quad (12)$$

With this  $H$  becomes

$$H = \frac{S^2 U_o^2}{x^2 D_e K_d \rho_p} \quad (13)$$

It is interesting to note that it is not necessary to know the actual fissure width, nor the actual water velocity for strongly volume sorbing nuclides. For the applications of interest in this study the time  $t$  is very much larger than  $Rt_w$ . For such cases the entity  $H$  is sufficient to describe the fissure.

Equation 9 is used to determine the concentration as it evolves with time in a single fissure.

#### Channeling model

The bedrock is assumed to consist of independent channels where the water flows with different velocities and flowrates. The model is based on the following concepts.

#### Derivation of model

At the inlet end of the channels a tracer is introduced. This is done simultaneously in all channels. At some distance downstream, the fluid from all channels is collected and mixed. The concentration of the mixture is measured over time. As some fissures carry the tracer faster than others, a step at the inlet will have spread when observed at the outlet. The fissure width distribution is  $f(\delta)$ . In a parallel walled fissure of width  $\delta_i$  with laminar flow, the flow rate  $Q(\delta_i)$  is proportional to the fissure width to the third power

$$Q(\delta_i) = k_1 \delta_i^3 l \quad (14)$$

where  $l$  is the breadth of the fissure. The velocity is proportional to the fissure width squared

$$U_i = k_1 \delta_i^2 \quad (15)$$

The residence time in fissures with width  $\delta_i$  over a given distance  $x$  is:

$$t_i = \frac{x}{k_1 \delta_i^2} \quad (16)$$

If a step with concentration  $c_0$  is introduced at the inlet of the set of fissures, it will travel the distance  $x$  in time  $t_i$  in fissures of width  $\delta_i$ . The fissures with residence times less than  $t$  will carry tracer, a fissure with residence time longer than  $t$  will carry no tracer.

The concentration obtained at the outlet end, at a time  $t$  when the effluent from all fissures is collected and mixed, is the sum of the flow from all fissures with tracer at the outlet, divided by the total flow.

$$\frac{c(t)}{c_0} = \frac{\int_0^{\infty} f(\delta)Q(\delta) d\delta}{\int_0^{\infty} f(\delta)Q(\delta) d\delta} = \frac{Q_t}{\bar{Q}} \quad (17)$$

$t$  is the residence time in fissure  $\delta^1(t)$  given by equation 16.

The above expression says that the flow  $Q_t$  from the tracer carrying fissures with widths  $\delta^1(t) \leq \delta < \infty$  is diluted by the total flow of water  $\bar{Q}$  from all fissures.

In general, for channels with a concentration break-through curve  $c(\delta, t)$  at the outlet, the concentration of the mixed effluent from all channels is

$$\frac{c(t)}{c_0} = \frac{\int_0^{\infty} f(\delta)Q(\delta)c(\delta, t)d\delta}{\int_0^{\infty} f(\delta)Q(\delta)d\delta} \quad (18)$$

Snow (1970) obtained the fissure width frequencies  $f(\delta)$  for various consolidated rocks including granites. Snow used data from water injection tests in boreholes and from direct measurements of fissure widths. He found the distribution to be log normal

$$f\left(\frac{\delta}{\mu}\right) = \frac{1}{A} \cdot e^{-\frac{1}{2}\left(\frac{\log \frac{\delta}{\mu}}{\sigma_l}\right)^2} \quad (19)$$

Snow (1970) found standard deviation varying between 0.057 and 0.394. His average value is 0.221. If a nonreacting and non-diffusing tracer were used to monitor the spreading of a tracer pulse in a system of fissures such as in the model above, an equivalent dispersion could be determined. This has been done by determining the second statistical moment (Neretnieks 1981).

For  $\sigma_{\rho} = 0.22$  the Peclet number which is defined below, is 1.1

$$\text{Pec} = \frac{U_f x}{D_L} \quad (20)$$

This indicates a very large dispersion. Such large dispersivities have been found in field experiments.

The channeling model indicates that the dispersion coefficient should increase with observation distance. This is usually found in field experiments. (Lallemand-Barrès and Peauderf 1978)

### 3. NUCLIDE RELEASE RATE

The time for canister penetration  $t_0$  is counted from an arbitrary starting time. It may be the time when the fuel is taken out from the reactor. At that time the waste contains  $N_{oi}$  Curies of nuclide  $i$ /ton of waste. The inventory of this nuclide will decay.

At time  $t$  the waste contains

$$N_i = N_{oi} \cdot e^{-\lambda_i t} \quad |\text{Ci/ton}| \quad (21)$$

In this single nuclide migration study only the part of the nuclide inventory originally present in the waste is considered. The release of nuclides is taken to be proportional to the dissolution rate of the waste. The dissolution rate is assumed to be constant over the whole dissolution time  $\Delta t$ . The release of a nondecaying nuclide during the dissolution time then is

$$n_{oi} = \frac{N_{oi}}{\Delta t} \quad \left| \frac{\text{Ci}}{\text{ton of original waste} \cdot \text{year}} \right| \quad (22)$$

At time  $t$  the release rate for a decaying nuclide then is

$$n_i = n_{oi} \cdot e^{-\lambda_i t} = \frac{N_{oi} \cdot e^{-\lambda_i t}}{\Delta t} \quad (23)$$

This amount of nuclide is diluted in a given stream of water which flows through the bedrock. Its initial concentration at  $t = 0$  will be  $c_{oi}$ . The nuclide travelling with the water will be subject to decay and loss due to diffusion into the matrix where the nuclide will have additional time to decay.

The concentration in the flowing water is observed at a distance  $x$  downstream. This concentration is obtained from equation (9) for a single channel and from equations (9, 18, 19) for the multiple channel case. The flowrate of nuclide  $i$  at distance  $x$  is

$$n_i^x = n_{oi} \cdot c_{fi}/c_{oi} = \frac{N_{oi}}{\Delta t} \cdot \frac{c_{fi}}{c_{oi}} \left| \frac{Ci}{\text{ton} \cdot \text{year}} \right| \quad (24)$$

The fraction of the original inventory of nuclide  $i$  arriving each year at distance  $x$  is

$$\frac{n_i^x}{N_{oi}} = \frac{1}{\Delta t} \cdot \frac{c_{fi}}{c_{oi}} \left| \frac{\text{fraction}}{\text{year}} \right| \quad (25)$$

## 4. DATA USED

The nuclides treated in this study are listed in table 1. Half-lives and mass sorption coefficients are taken from the Nuclear Fuel Safety project study (KBS 1978). Mass sorption coefficients are converted to volume sorption coefficients by multiplying it with the rock density  $\rho_p = 2700 \text{ kg/m}^3$ .

The data needed for the present model are

Time of canister penetration	$t_o$	year
Time for dissolution of waste <sup>a</sup>	$\Delta t$	year
Standard deviation of fissure width (channel) distribution	$\sigma_l$	—
Matrix penetration parameter eq (10) or (13) for nuclide $i$	$H_i$	year <sup>-1</sup>

a) constant dissolution rate is assumed.

Times for canister penetration of 0.1, 40, 5000 and  $10^5$  years have been used. Dissolution times of 30 000, 300 000, 500 000,  $3 \cdot 10^6$  and  $10^9$  have been used. The standard deviation of the fissure width distribution  $\sigma_l$  has been given the values 0 (equal fissure widths) and 0.221.

The matrix penetration parameter  $H$  includes many entities

$$H_i = \frac{S_o^2 U_o^2}{x^2 D_e K_d \rho_p} \quad (13)$$

With  $U_o = K_p i$  and with a hydraulic conductivity  $K_p$  of  $10^{-9}$  m/s and hydraulic gradient  $i = 0.01$  m/m  $U_o = 10^{-11} \frac{m^3}{m^2} \cdot s$  or approximately  $0.3$  l/m<sup>2</sup> · year. With a fissure spacing  $S = 1$  m and an effective diffusivity  $D_e = 10^{-12}$  m<sup>2</sup>/s, the group  $\alpha$

$$\alpha = \frac{S^2 \cdot K_p^2 \cdot i^2}{D_e} \quad (26)$$

has a value of  $10^{-10}$  |m<sup>2</sup>/s|.

The same value of  $\alpha$  could be obtained with another combination of values for the individual variables. The noted values of  $S$ ,  $K_p$ ,  $i$  and  $D_e$  are in the range of values expected for the Swedish bedrock. They are used as central case values.

The water velocity which gives the water residence time  $t_w$  is calculated from equation (16). The constant  $k_1$  is obtained from laminar flow theory applied to flow between infinite parallel plates (Snow 1968).

$$k_1 = g/12 \nu i$$

where the gravitational constant  $g = 9.81$  m/s<sup>2</sup> and the viscosity of water  $\nu = 10^{-6}$  m<sup>2</sup>/s. The water residence time  $t_w$  per se has no influence of the results for all practical purposes. This applies also to non-interacting nuclides such as I-129 except for the earliest breakthrough times if the assumption (equation 5) is valid that the slab of rock between two fissures will never be saturated.

## 5. COMPUTED CASES

Table 2 gives the data for the cases presented in this paper. The cases are divided in two main groups. In the first, all the fissures have equal width which implies that there is no channeling and no hydrodynamic dispersion. These are the cases 1-8. The second group assumes a strong channeling with  $\sigma_g = 0.221$ .

## 6. PRESENTATION OF RESULTS

For every case the effluent fraction  $n_i^x/N_{oi}$  of each nuclide have been calculated at various distances  $x$  and for times up to a maximum of  $10^9$  years. Figures 1-8 show the breakthrough curves at 1000 m distance for all those nuclides which have a  $c_f/c_o$  larger than  $10^{-9}$  at any time.

It can be seen that in case 1 with no channeling only I-129 will arrive with a release rate higher than  $10^{-13}$  of the original inventory per year (Figure 1). The same case with severe channeling is shown in figure 4 (case 13).

Another type of plot is shown in figures 9-26. They have been constructed by plotting the maximum points in figures 1-8 (and many more) versus distance. They thus show the maximum fraction of the original content of the inventory which can reach a certain distance at any time. At all other times the fraction will be less (or equal).

## 7. DISCUSSION

The hydraulic conductivity  $K_p$ , hydraulic gradient  $i$  and the fissure spacing  $S$  influence the results in the same way. All three parameters are part of the group

$$\alpha = \frac{S^2 \cdot K_p^2 \cdot i^2}{D_e} \quad (26)$$

The effective diffusivity is also included in this group.

The four entities influence the results only through this group and need thus not be considered separately.

There is an inherent assumption which may not be valid when the fissure spacing is small or when the nuclide sorbs poorly (small  $K_{dp}$ ). The assumption is that the penetration depth of the nuclide into the rock from the surface of the fissure is smaller than half the distance between fissures. Penetration depths for various  $K_{dp}$  values have been calculated (Neretnieks 1980).



For  $K_d \rho_p$  values around  $100 \text{ m}^3/\text{m}^3$  (Cs, Sr, Tc) about  $5 \cdot 10^4$  years will suffice to penetrate 0.5 and about  $1.2 \cdot 10^8$  will be needed to penetrate 25 m. For radium and the actinides with  $K_d \rho_p \sim 10^3$  (Pa, U, Pu) it takes 10 times longer to penetrate the same depths and for Th, Am and Cm with  $K_d \rho_p \sim 10^4$  and larger it will take  $\sim 5 \cdot 10^6$  and  $1.2 \cdot 10^{10}$  years to penetrate 0.5 and 25 m respectively. Iodide with  $K_d \rho_p = 0.005$  will need  $\sim 2.5$  years to penetrate 0.5 m and 6200 years to penetrate 25 m.

The analysis will not be valid for iodide except for the very short times. The presented curves will underestimate the effluent concentration for iodide. For most of the other nuclides the larger fissure spacing 50 m will not infringe upon the assumption whereas a smaller fissure spacing 1 m will invalidate the results for Cs-135 and Tc-99 for a high hydraulic conductivity ( $K_p = 10^{-7} \text{ m/s}$  fig 3 and figure 12). For smaller hydraulic conductivities,  $10^{-8}$ , these nuclides will become unimportant at distances over 1000 m. It is very doubtful, however, if fissures with equal widths and hydraulic conductivities may be spaced as closely as 1 m. The cases with equal fissure widths ( $\sigma_l = 0$ ) should more be seen as cases for single large fissures. The fissure spacing S would then mean that if such fissures were placed at distances S the average permeability of such a rock mass would become  $K_p$ .

For a rock mass with varying fissure widths (and thus conductivities) the faster fissures will dominate the breakthrough curves. As these larger fissures make up a small fraction of all fissures they will be much more widely spaced than S. The easy and more interesting part of the breakthrough curves may thus not seriously violate the assumption.

The results from these calculations can be compared to those in KBS 1978 concerning the direct deposition of spent fuel. Case 21 has the same  $t_0$  and  $\Delta t$  and sorption coefficients  $K_d$  as the main case in KBS 1978. Practically the same water flowrate is used,  $U_0 = 3 \cdot 10^{-4} \text{ m}^3/\text{m}^2 \cdot \text{year}$  in case 21 and  $2 \cdot 10^{-4} \text{ m}^3/\text{m}^2 \cdot \text{year}$  in KBS 1978. The fissure spacing is the same  $S = 1 \text{ m}$ . The water velocity as such does not influence the results for the sorbing nuclides. The distance is not defined in KBS 1978 but is much larger than 1000 m.

This type of comparison is quite misleading, however. In KBS 1978 the computed retardation factors were based on the assumptions of surface sorption. The retardation factor  $R_i$  was calculated from

$$R_i = 1 + \frac{a K_{ai}}{\epsilon_f} (1 - \epsilon_f)$$

where

$$a = \text{specific surface} \left| \frac{\text{m}^2}{\text{m}^3 \text{ rock}} \right|$$

$$K_{ai} = \text{surface sorption coefficient} \quad |m|$$

$$\epsilon_f = \text{flow porosity of bedrock}$$

The flow porosity of the bedrock was taken to be  $10^{-5}$ . The use of this porosity in the present study would lead to an equivalent migration path length of 60 000 m instead of 1 000 m to obtain the residence time of 3 000 years used in (KBS 1978). Under such conditions the present model would predict that no nuclide except I-129 would reach the biosphere in significant amounts.

It should also be noted that the impact of channeling is severe. A comparison of case 1 and 13 shows that in case 1 (figures 1 and 9) only I-129 and U-238 travel past a 100 m mark in any appreciable amount. In this case the fissures are assumed to be of equal width and no dispersion is assumed (In KBS 1978 the dispersion was for all practical purposes neglected).

With severe channeling - case 13 - figure 23 shows that practically all the nuclides studied will be observable at the 100 m point. Even at a distance of 1 000 m, 9 nuclides will arrive with a release larger than  $10^{-14}$ /year of the original inventory.

## 8. USE OF RESULTS

The results in this study can be used to assess the influence of some important entities on the migration of the most important radionuclides. They may also under some circumstances be used to indicate what effluent rates to the biosphere to expect. The possible combinations of input-parameter values are vast and only a few selected combinations were used. The initial canister breakthrough time  $t_0$  was taken to be short in most cases in order to include the shortlived nuclides. Any case can be recalculated to another  $t_0$  by accounting for the decay. This is done by multiplying the release by  $e^{-\lambda_i(t_0^2 - t_0^1)}$  for every individual nuclide  $t_0^2$  is the new breakthrough time and  $t_0^1$  is the time for which the results are available.

Cases 1-8 apply to no channeling or single fissure cases. They may be used to indicate the release from an individual fissure. One application would be to study transport in an odd large fracture. If the  $K_p$ ,  $i$ ,  $S$  and  $D_e$  values for the case of interest can be found in table 2, the results can be applied in a straightforward manner. When the  $K_p$ ,  $i$ ,  $S$ , and  $D_e$  values cannot be found the combination  $\alpha$  (see equation 26) is determined, and the cases having  $\alpha$  nearest to the desired case are used.

As an example consider a case where a fissure is expected at every 100 m. The bedrock with these fissures has an average hydraulic conductivity  $K_p = 10^{-9}$  m/s. In this rock a 2 m straddle packer test would give a section with  $K_p = 0.5 \cdot 10^{-7}$  m/s when the fissure was found, assuming that the rest of the rock is tight. For  $i = 0.003$  m/m and  $D_e = 10^{-13}$  m/s we obtain  $\alpha = 0.910 \cdot 10^{-6}$  m<sup>2</sup>/s. In table 3 we find that case 3 is nearest,  $\alpha = 10^{-6}$  m<sup>2</sup>/s. For a leach time of  $3 \cdot 10^4$  years figures 3 and 12 can be used.

For the single fissure case it is very easy to calculate the breakthrough using the equation (9). It is easily done using a pocket calculator and a table of values of the error function.

When channeling is to be included, cases 13-25 can be used in the same way as exemplified for the single channel case. As can be seen by comparing cases 1 and 13 or 4 and 17, 16 and 20 etc. it is evident that for the same average hydraulic conductivity of the bedrock, same fissure spacing etc., a variation of fissure widths will lead to a strong increase in release rates.

The channeling parameter chosen  $\sigma_l = 0.221$  probably is on the high side for large distances. There is, however, no experimental evidence at present to really assess the channelling or dispersion mechanisms over large distances in fissured bedrock.

Case 21, figures 7 and 23, has about the same basic values of  $S$ ,  $K_p$  and  $i$  as those used in the KBS II (1978) safety analysis for spent fuel. The travel distance was not defined in that study.

Table 3 gives a comparison between the KBS II (1978) results and the results from case 21 for 1000 m and 5000 distances. The inventory of radionuclides in  $10^4$  tonnes of  $10^5$  years old spent fuel as given in table 8-2 in KBS II 1978 is used for the calculations. Iodide is not included as the calculations do not apply for a non-sorbing nuclide at these very long contact times.

Whereas the KBS II 1978 study predicted releases in the mCi range for some nuclides, this model predicts maximum releases of at most some 10:s of  $\mu$ Ci. It should be noted that in the present model a very strong channeling has been assumed. This was not included in KBS II (1978).

Nuclide	Halflife	Volume sorption coefficient
	$T_{1/2}$ years	$K_d \rho_p$ <sup>a)</sup> $m^3 / m^3$
Sr-90	28.8	43
Tc-99	212 000	135
I-129	$17 \cdot 10^6$	0.005 <sup>b)</sup>
Cs-135	$3 \cdot 10^6$	170
Cs-137	30.2	170
Ra-226	1 600	1 350
Th-229	7 340	6 480
Th-230	80 000	6 480
Pa-231	32 500	1 620 <sup>c)</sup>
U-233	162 000	3 240
U-234	247 000	3 240
U-238	$4.51 \cdot 10^9$	3 240
Np-237	$2.14 \cdot 10^6$	3 240
Pu-238	86	810
Pu-239	24 400	810
Pu-240	6 580	810
Pu-241	13.2	810
Pu-242	379 000	810
Am-241	458	86 000
Am-242-m	152	86 000
Am-243	7 370	86 000
Cm-244	17.6	43 000 <sup>d)</sup>
Cm-245	9 300	23 000

Table 1. Nuclides considered in this study.  
No chain decay is accounted for.

a) Nuclear fuel safety project (1978), vol.II  
Technical report, Stockholm.

b)  $K_d \rho_p$  equal to porosity of granite matrix.

c) Assumed half that of U.  
Calculated only for case 13.

d) See J.Geophys.Res. 85, 1980, p 4379.  
Half of value for Am-243

Case	No channeling $\sigma_\ell = 0$								Channeling $\sigma_\ell = 0.221$								
	1	2	3	4	5	6	7	8	13	16	17	18	20	21	23	24	25
$\alpha$ m <sup>2</sup> /s	$10^{-10}$	$10^{-4}$	$10^{-6}$	$10^{-8}$	$10^{-10}$	$10^{-10}$	$10^{-10}$	$10^{-10}$	$10^{-10}$	$10^{-8}$	$10^{-12}$	$10^{-10}$	$10^{-10}$	$10^{-10}$	$2.2 \cdot 10^{-10}$	$2.2 \cdot 10^{-8}$	$2.2 \cdot 10^{-6}$
$K_p$ m/s	$10^{-9}$	$10^{-6}$	$10^{-7}$	$10^{-8}$	$10^{-9}$	$10^{-9}$	$10^{-9}$	$10^{-9}$	$10^{-9}$	$10^{-8}$	$10^{-10}$	$10^{-9}$	$10^{-9}$	$10^{-9}$	$10^{-10}$	$10^{-9}$	$10^{-8}$
$i$ m/m	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.003	0.003	0.003
$S$ m	1	1	1	1	1	1	1	1	1	1	1	1	1	1	50	50	50
$t_o$ years	40	40	40	40	5000	5000	5000	40	40	40	40	5000	5000	$10^5$	0.1	0.1	0.1
$\Delta t$ years	$3 \cdot 10^4$	$3 \cdot 10^4$	$3 \cdot 10^4$	$3 \cdot 10^4$	$3 \cdot 10^4$	$3 \cdot 10^5$	$3 \cdot 10^6$	$10^9$	$3 \cdot 10^4$	$3 \cdot 10^4$	$3 \cdot 10^4$	$3 \cdot 10^4$	$3 \cdot 10^6$	$5 \cdot 10^5$	$10^9$	$10^9$	$10^9$
Figure no:	1,9	2,10,11	3,12	1,13	14	15	16	17	4,18	19	20	5,21	6,22	7,23	8,24	25	26

$D_e = 10^{-12}$  m<sup>2</sup>/s for all cases

Table 2. Data for the cases presented.  
Cases 1 and 13 are the central cases without and with channeling.

Nuclide	Radioactivity per $10^4$ tonnes after $10^5$ years	Maximum inflow to recipient area		
		Case 21 1000 m Ci/yr	Case 21 5000 m Ci/yr	KBS II (1978) Ci/yr
Cs-135	2 500	$3 \cdot 10^{-6}$	$8 \cdot 10^{-8}$	$3 \cdot 10^{-4}$
Tc -99	100 000	$3 \cdot 10^{-5}$	$5 \cdot 10^{-7}$	$2 \cdot 10^{-5}$
U-238	3 100	$3 \cdot 10^{-7}$	$9 \cdot 10^{-9}$	$6 \cdot 10^{-3}$
Pu-242	13 000	$10^{-6}$	$1.3 \cdot 10^{-8}$	$\ll 10^{-9}$
Np-237	11 000	$3 \cdot 10^{-7}$	$6 \cdot 10^{-9}$	$10^{-10}$
U-234	14 000	$10^{-7}$	$8 \cdot 10^{-10}$	$6 \cdot 10^{-3}$
U-233	3 800	$2 \cdot 10^{-8}$	$2 \cdot 10^{-10}$	$10^{-10}$
Th-230	9 000	$2 \cdot 10^{-9}$	$3 \cdot 10^{-11}$	$3 \cdot 10^{-3}$
Pu-239	200 000	$2 \cdot 10^{-8}$	$2 \cdot 10^{-10}$	n.a.
Ra-226	1 600	n.a.*	n.a.*	$3 \cdot 10^{-3}$

\* Ra-226 is a daughter of Th 230 and will in these circumstances be very near radioactive equilibrium with Th-230. The Ra-226 activity would then be of the same magnitude as Th-230 activity.

Table 3. Maximum inflow of activity to recipient area from a repository with 10 000 tonnes of spent fuel. Case 21  
 $K_p = 10^{-9}$  m/s,  $i = 0.01$  m/m,  $D_e = 10^{-12}$  m<sup>2</sup>/s,  $S = 1$  m,  
 $t_o = 10^5$  years,  $\Delta t = 5 \cdot 10^5$  years.

## NOTATION

a	specific surface	$\text{m}^2/\text{m}^3$ fluid
c	concentration in the liquid	$\text{mol}/\text{m}^3$
$c_0$	initial concentration in the liquid	$\text{mol}/\text{m}^3$
$c_f$	concentration in the liquid in a fissure	$\text{mol}/\text{m}^3$
$c_p$	concentration in the liquid in a pore	$\text{mol}/\text{m}^3$
$c_s$	concentration on the solid	$\text{mol}/\text{m}^2$
$D_a$	apparent diffusivity $D_a = D_e/K_d \rho_p$	$\text{m}^2/\text{s}$
$D_e$	effective diffusivity $= D_e \epsilon_p$	$\text{m}^2/\text{s}$
$D_L$	dispersion coefficient	$\text{m}^2/\text{s}$
$D_p$	diffusivity in micropores	$\text{m}^2/\text{s}$
G	see equation 8	
g	gravitational constant	$\text{m}/\text{s}^2$
H	see equations 10 and 13	
i	hydraulic gradient	$\text{m}/\text{m}$
$K_a$	surface sorption coefficient	$\text{m}$
$K_d \rho_p$	volume sorption coefficient	$\text{m}^3/\text{m}^3$
$K_p$	hydraulic conductivity	$\text{m}/\text{s}$
n	nuclide release rate at repository	$\text{Ci}/\text{ton}, \text{year}$
$n^x$	nuclide release rate at distance x	$\text{Ci}/\text{ton}, \text{year}$
R	retardation factor due to surface sorption	-
Q	flow rate	$\text{m}^3/\text{s}$
$\bar{Q}$	mean flow rate	$\text{m}^3/\text{s}$
$Q_t$	flow rate carrying tracer	$\text{m}^3/\text{s}$
R	retardation factor	-
S	fissure spacing	$\text{m}$
t	time	$\text{s}$
$t_0$	time at which leaching of waste starts	$\text{s}$
$t_w$	water residence time	$\text{s}$
$\Delta t$	time for dissolution of waste	$\text{s}$
$U_f, U$	fluid velocity in a fissure	$\text{m}/\text{s}$
$U_0$	water flux	$\text{m}^3/\text{m}^2$
x	distance in flow direction	$\text{m}$
z	distance into rock matrix	$\text{m}$
$\alpha$	see equation 26	-
$\delta$	fissure width	$\text{m}$



$\epsilon_f$	porosity considering fissure volume only	-
$\epsilon_p$	porosity of rock matrix excluding fissures	-
$\lambda$	radioactive decay constant	$s^{-1}$
$\mu$	mean fissure width	m
$\nu$	viscosity of fluid (water)	$m^2/s$
$\rho$	density of rock matrix	$kg/m^3$
$\sigma_l$	standard deviation of the logarithm of fissure widths	-

#### Subscript

i	nuclide i or fissure i
o	at time = 0

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## LITERATURE

Brace W F, Orange A S, Madden T R: The effect of pressure on the electrical resistivity of water-saturated crystalline rocks. J. Geophys. Res., 70, 1965, p 5669.

KBS - Nuclear fuel safety project: Handling of spent nuclear fuel and final storage of vitrified high-level reprocessing waste, vol IV, Safety analysis report, Stockholm, 1977.

KBS - Nuclear fuel safety project: Handling and final storage of unprocessed spent nuclear fuel, vol II. Technical report, Stockholm, 1978.

Lallemand Barrès A, Peaudecerf P: Recherche des relations entre la valeur de la dispersivité macroscopique d'un milieu aquifère, ses autres caractéristiques et les conditions de mesure. Bulletin du B.R.G.M. Section III, 4, 1978, p 277.

Larson S Å, Tullborg E L, Lindblom S: Sprickmineralogiska undersökningar. PRAV Report no 4.20, April 1981 (in Swedish).

Neretnieks I: Diffusion in the rock matrix: An important factor in radionuclide retardation? J. Geophys. Res., 85, 1980, p 4379.

Neretnieks I: Some difficulties in interpreting in-situ tracer tests. Paper presented at the third international symposium on the "Scientific basis for nuclear waste management" arranged by Materials Research Society, Boston, Nov. 16-20, 1980 b.

Neretnieks I, Eriksen T, Tähtinen P: Tracer Movement in a Single Fissure in Granitic Rock - Some Experimental Results and their Interpretation. Report no 4.21 1981. National Council for Radioactive Waste PRAV, Stockholm, 1981.

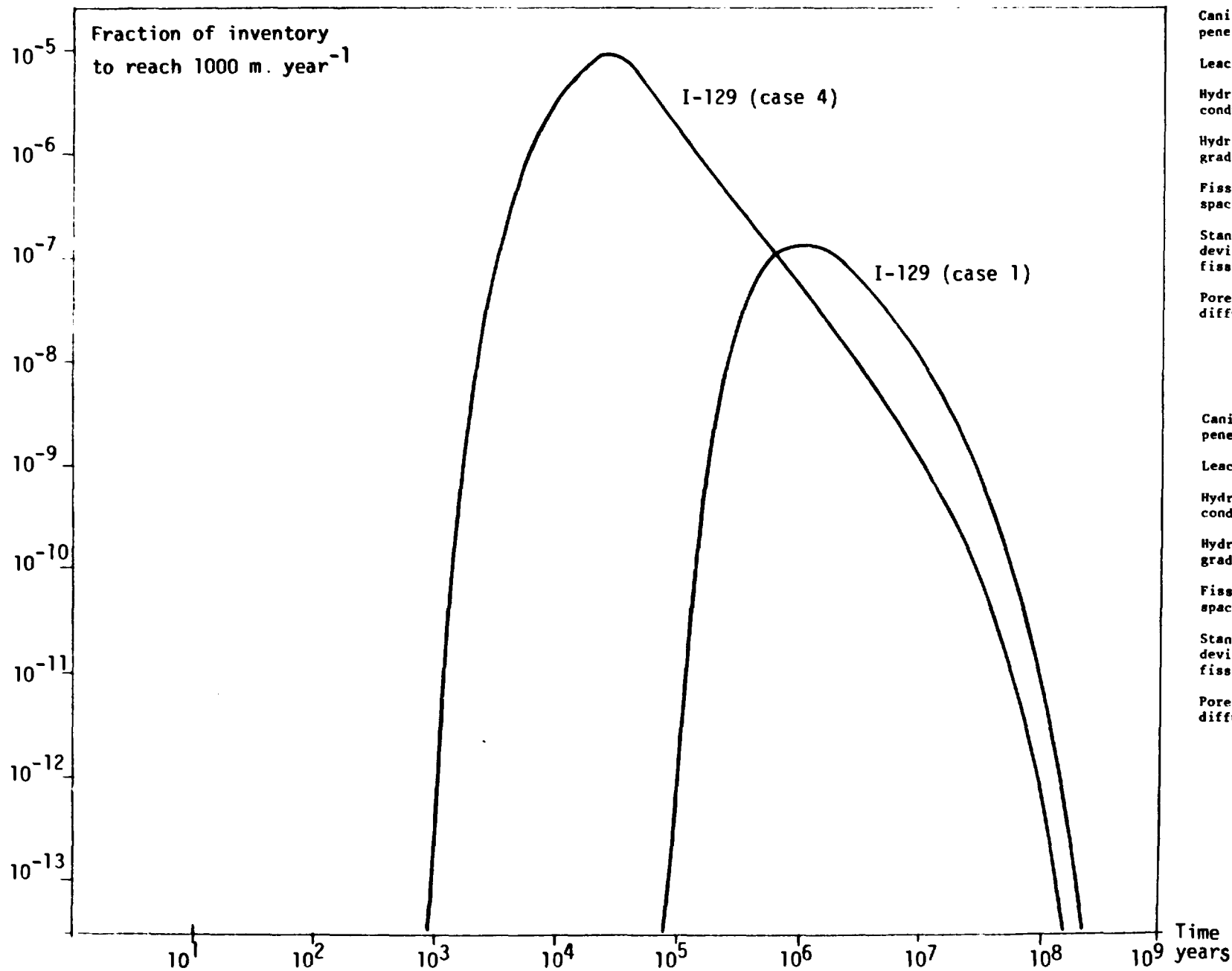
Neretnieks I: Prediction of Radionuclide Migration in the Geosphere. - Is the Porous Flow Model Adequate? International symposium on migration in the terrestrial environment of long lived radionuclides from the nuclear fuel cycle. IAEA symposium, Knoxville Tennessee, USA, 27-31 July, 1981.

Skagius C, Svedberg G, Neretnieks I: A Study of Strontium and Cesium Sorption on Granite. Report no 4.26 1981. National Council for Radioactive Waste PRAV, Stockholm, 1981.

Snow D T: Rock Fracture Spacings, Openings and Porosities. Journal of the Soil Mechanics and Foundations Division, Jan 1968, p 73. AICHE J 94, SMI p 73-91.

Snow D T: The Frequency and Apertures of Fractures in Rock. Int. J. Rock Mech. Min. Sci., 7, 1970, p 23-40.

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Canister penetration	$t_o$	40	years
Leach time	$\Delta t$	$3 \cdot 10^4$	years
Hydraulic conductivity	$K_p$	$10^{-9}$	m/s
Hydraulic gradient	$i$	0.01	m/m
Fissure spacing	$S$	1	m
Standard deviation fiss. width	$\sigma$	0	-
Pore diffusivity	$D_p^c$	$10^{-12}$	m <sup>2</sup> /s

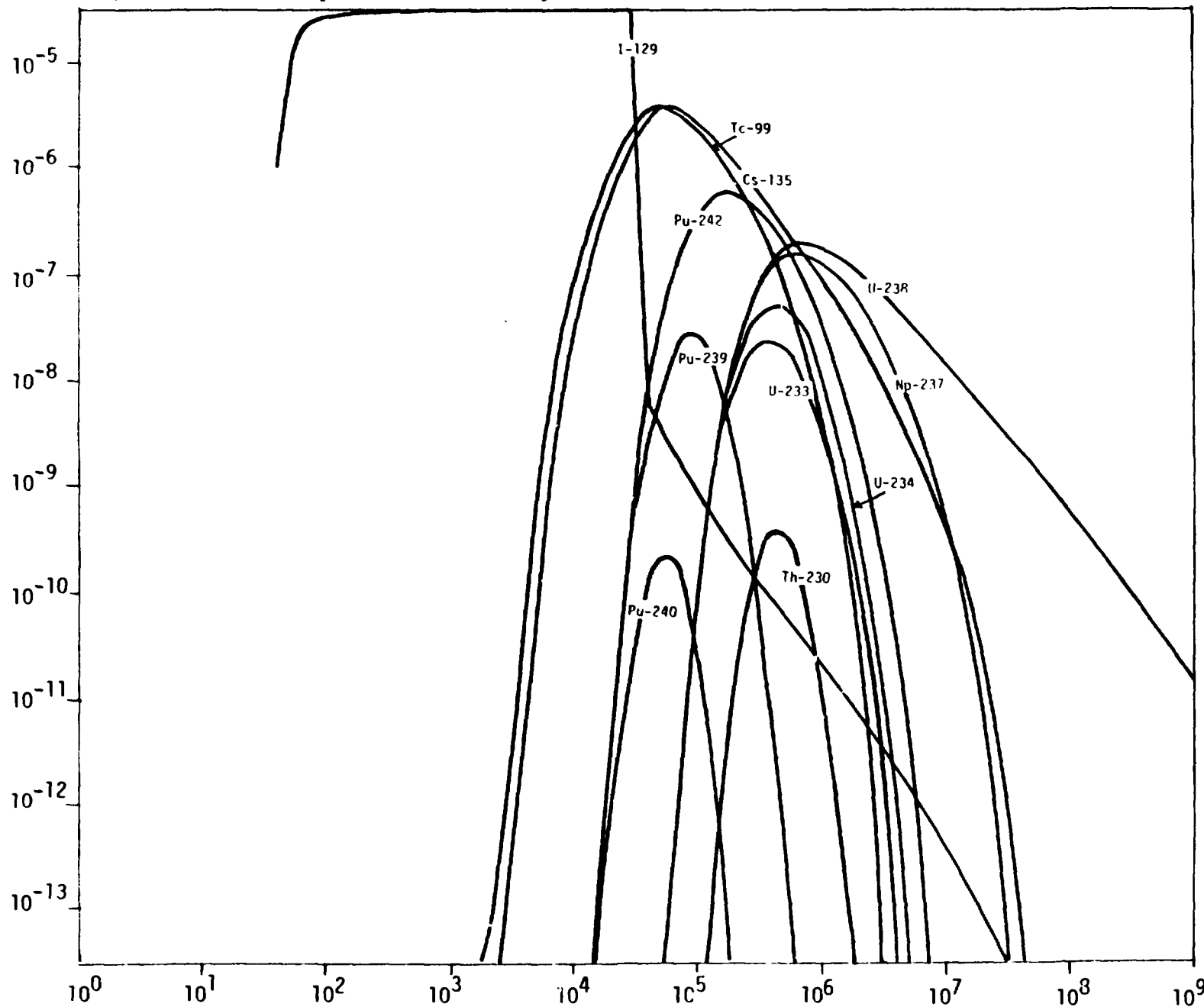
#### Case 1

Canister penetration	$t_o$	40	years
Leach time	$\Delta t$	$3 \cdot 10^4$	years
Hydraulic conductivity	$K_p$	$10^{-8}$	m/s
Hydraulic gradient	$i$	0.01	m/m
Fissure spacing	$S$	1	m
Standard deviation fiss. width	$\sigma$	0	-
Pore diffusivity	$D_p^c$	$10^{-12}$	m <sup>2</sup>

#### Case 4

Figure 1

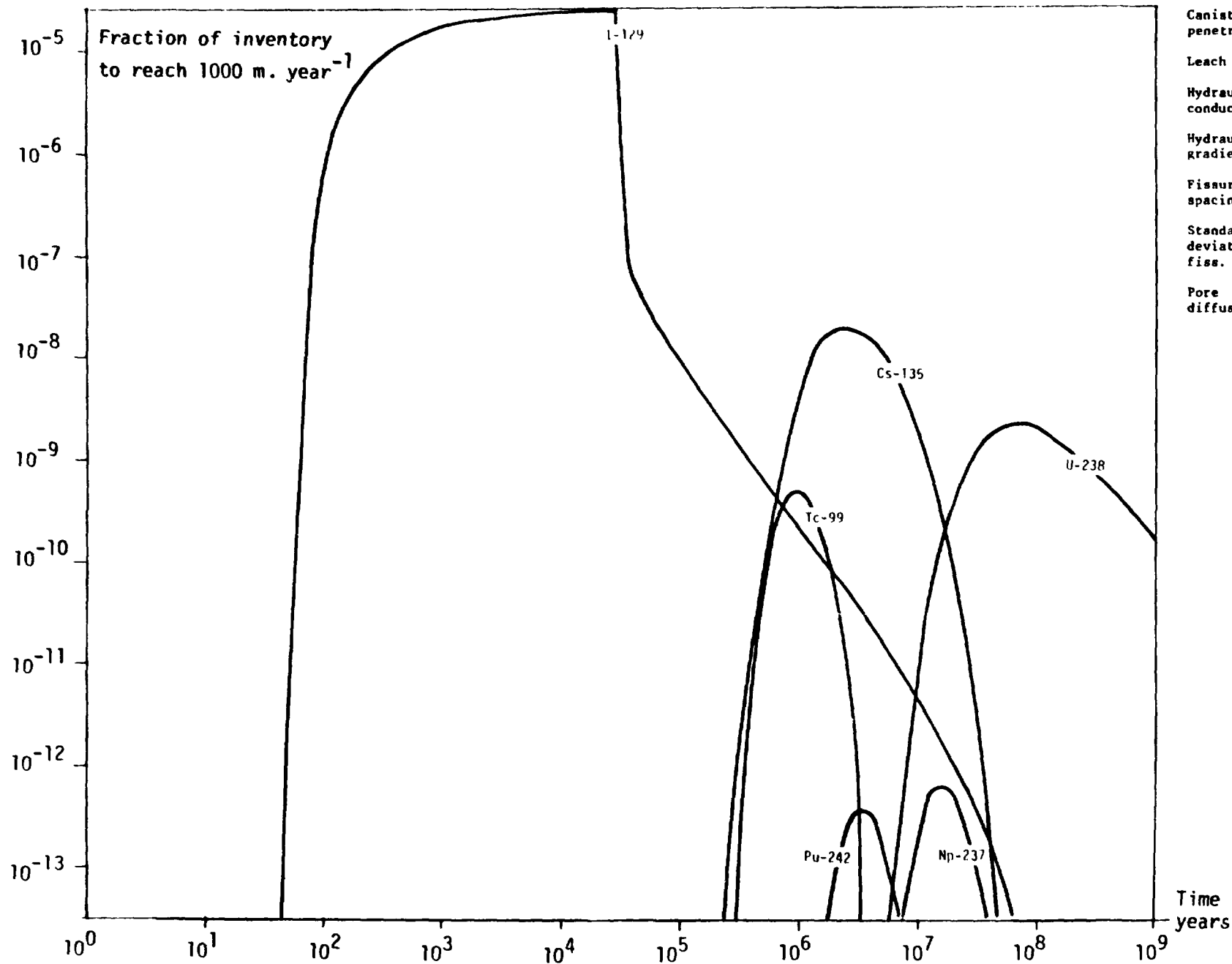
Fraction of inventory to reach 1000 m. year<sup>-1</sup>



Canister penetration	$t_0$	40	years
Leach time	$\Delta t$	$3 \cdot 10^4$	years
Hydraulic conductivity	$K_p$	$10^{-6}$	m/s
Hydraulic gradient	$i$	0.01	m/m
Fissure spacing	$S$	1	m
Standard deviation fiss. width	$\sigma$	0	-
Pore diffusivity	$D_p \epsilon_p$	$10^{-12}$	$m^2/s$

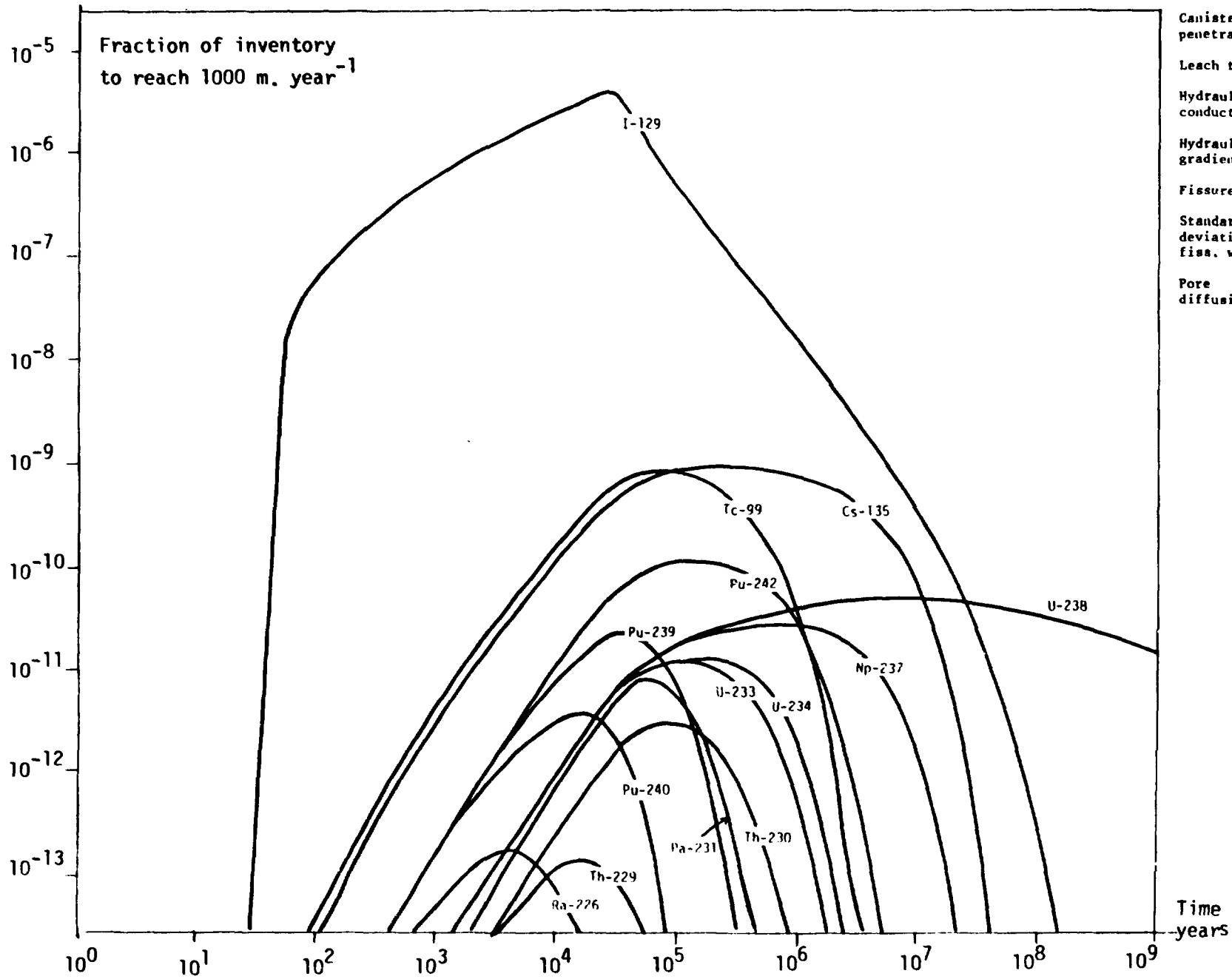
Time years

Figure 2



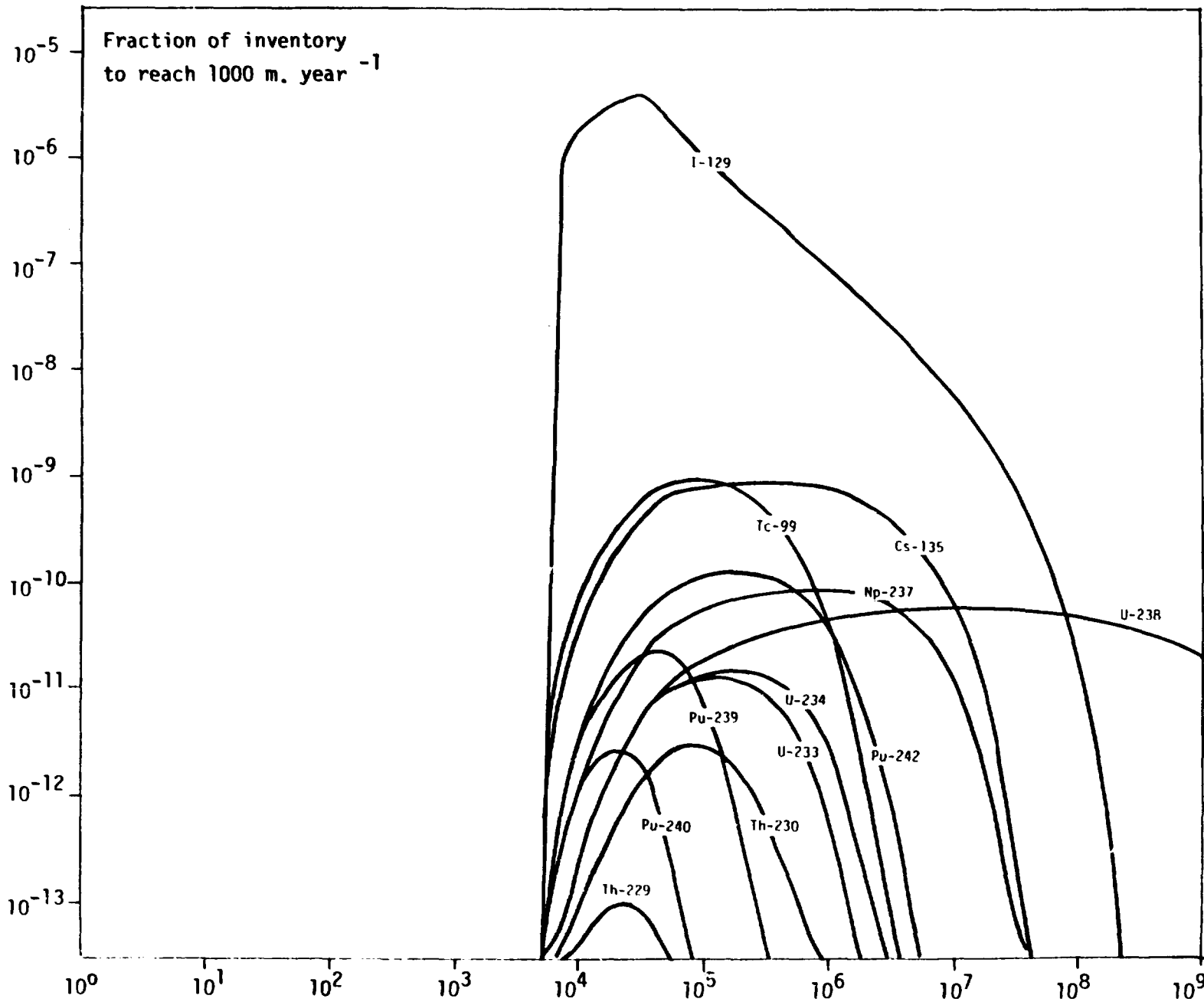
Canister penetration	$t_0$	40	years
Leach time	$\Delta t$	$3 \cdot 10^4$	years
Hydraulic conductivity	$\kappa_p$	$10^{-7}$	years
Hydraulic gradient	$i$	0.01	m/m
Fisure spacing	$S$	1	m
Standard deviation fiss. width	$\sigma$	0	-
Pore diffusivity	$D_{pP}^c$	$10^{-12}$	m <sup>2</sup> /s

Figure 3



Canister penetration	$t_n$	40	years
Leach time	$\Delta t$	$3 \cdot 10^4$	years
Hydraulic conductivity	$K_p$	$10^{-9}$	m/s
Hydraulic gradient	$i$	0.01	m/m
Fissure spacing	$S$	1	m
Standard deviation fiss. width	$\sigma$	0.221	-
Pore diffusivity	$D_{p p}^c$	$10^{-12}$	m <sup>2</sup> /s

Figure 4



Canister penetration	$t_0$	5000	years
Leach time	$\Delta t$	$3 \cdot 10^4$	years
Hydraulic conductivity	$K_p$	$10^{-9}$	m/s
Hydraulic gradient	$i$	0.01	m/m
Fissure spacing	$S$	1	m
Standard deviation fiss. width	$\sigma$	0.221	-
Pore diffusivity	$D_{p \epsilon_p}$	$10^{-12}$	m <sup>2</sup> /s

Figure 5



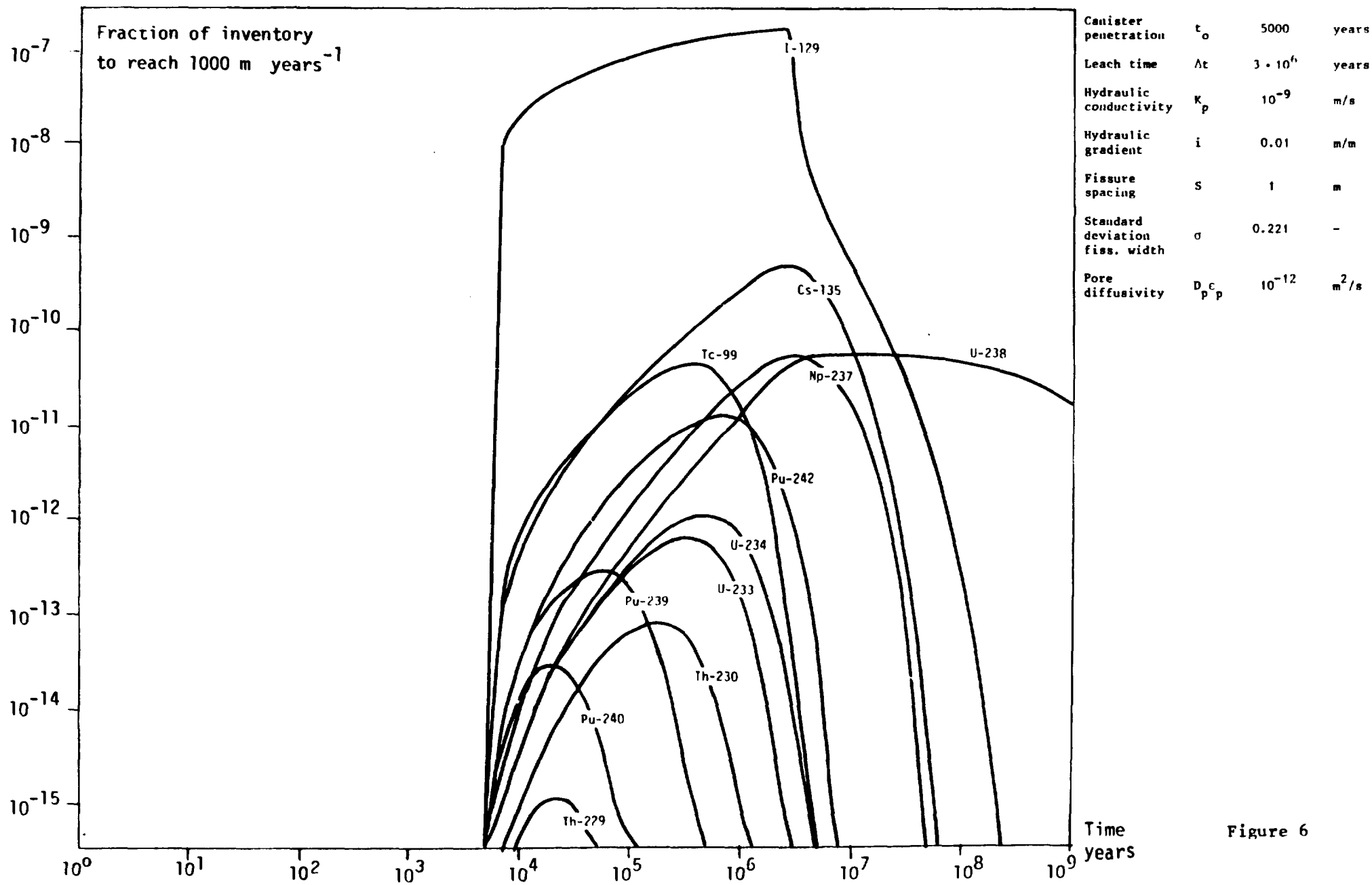
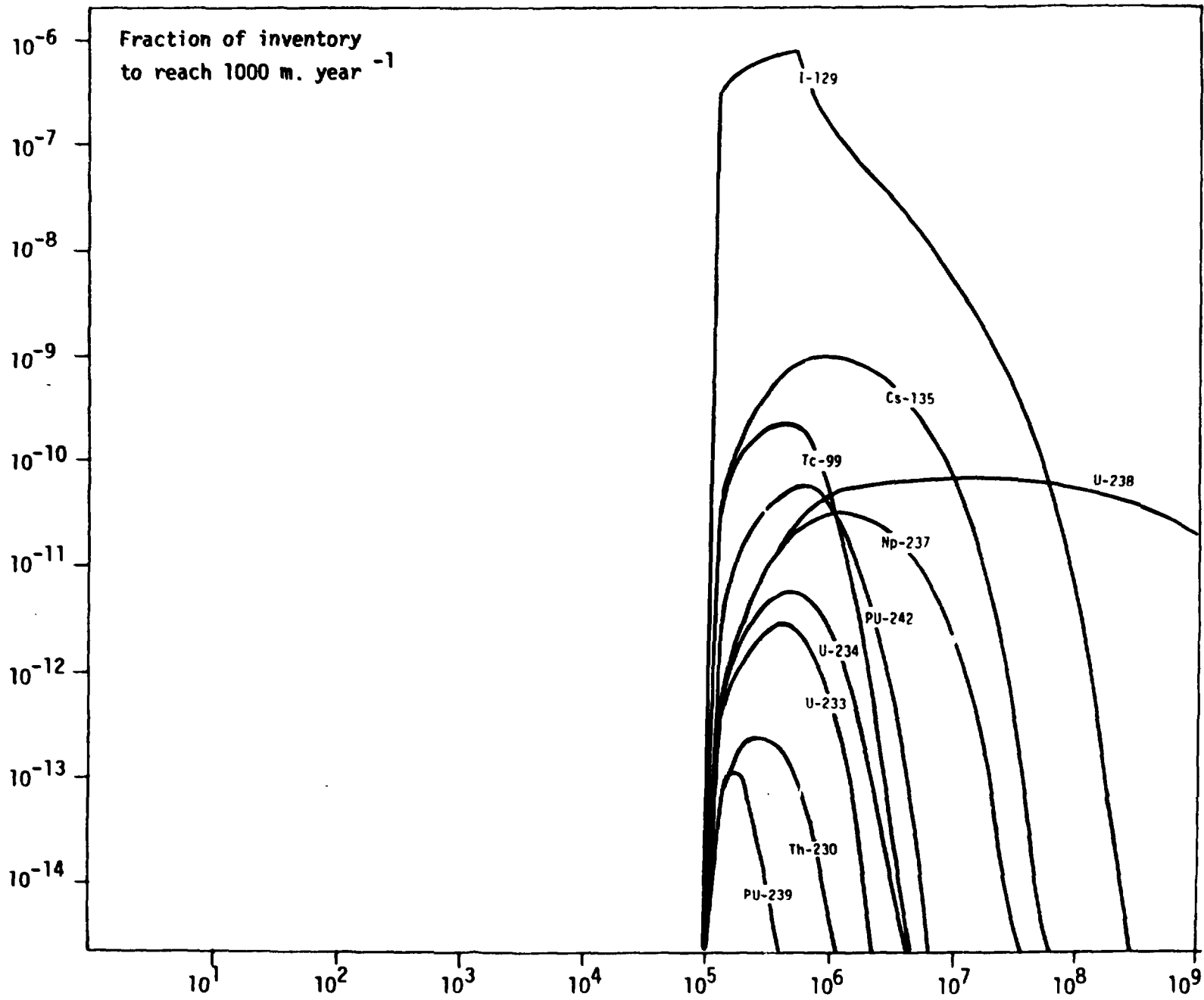


Figure 6



Canister penetration	$t_0$	$10^5$	years
Leach time	$\Delta t$	$5 \cdot 10^5$	years
Hydraulic conductivity	$K_p$	$10^{-9}$	m/s
Hydraulic gradient	$i$	0.01	m/m
Fissure spacing	$S$	1	m
Standard deviation fiss. width	$\sigma$	0.221	-
Pore diffusivity	$D_p^c$	$10^{-12}$	m <sup>2</sup> /s

Figure 7

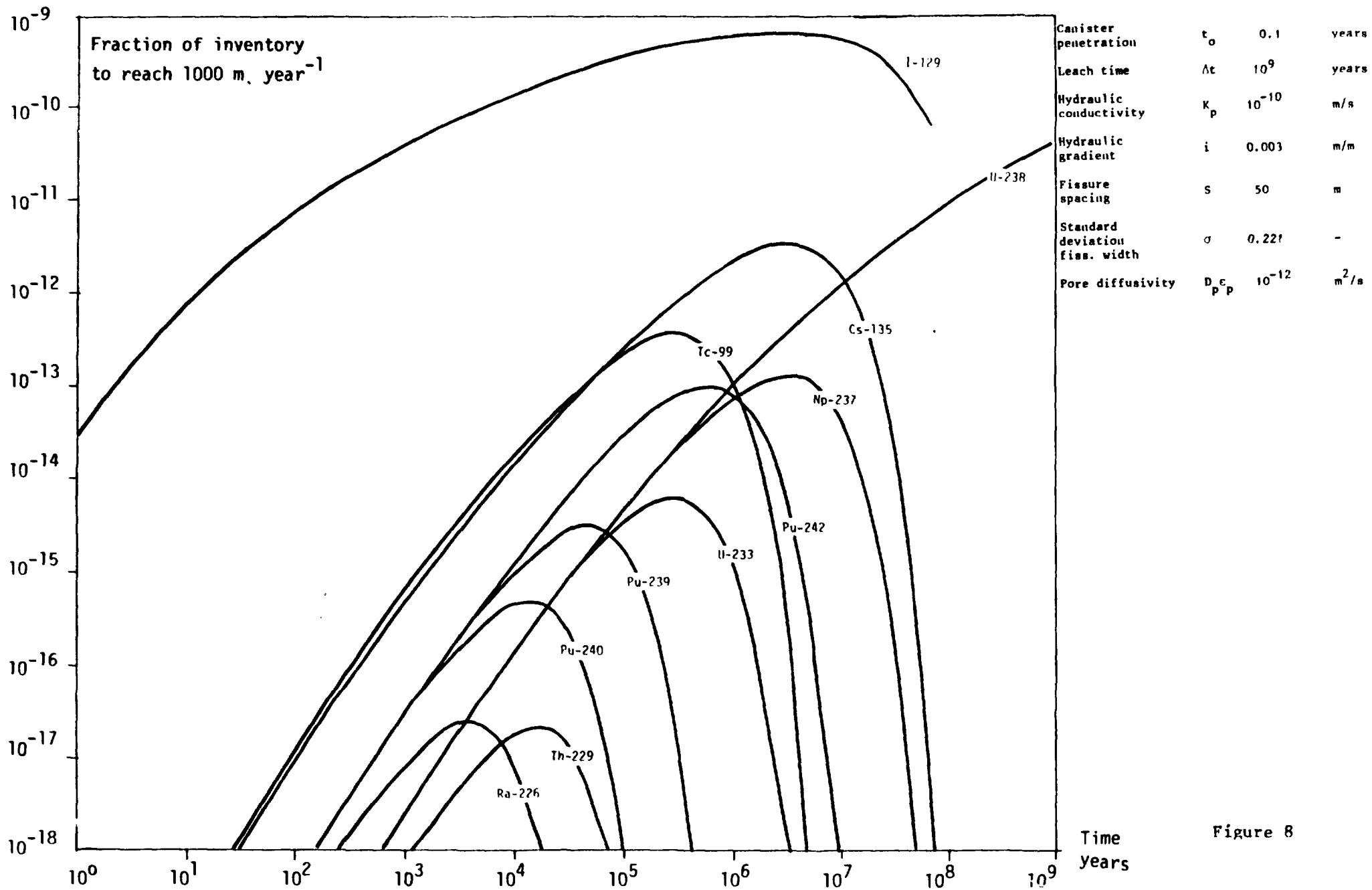


Figure 8

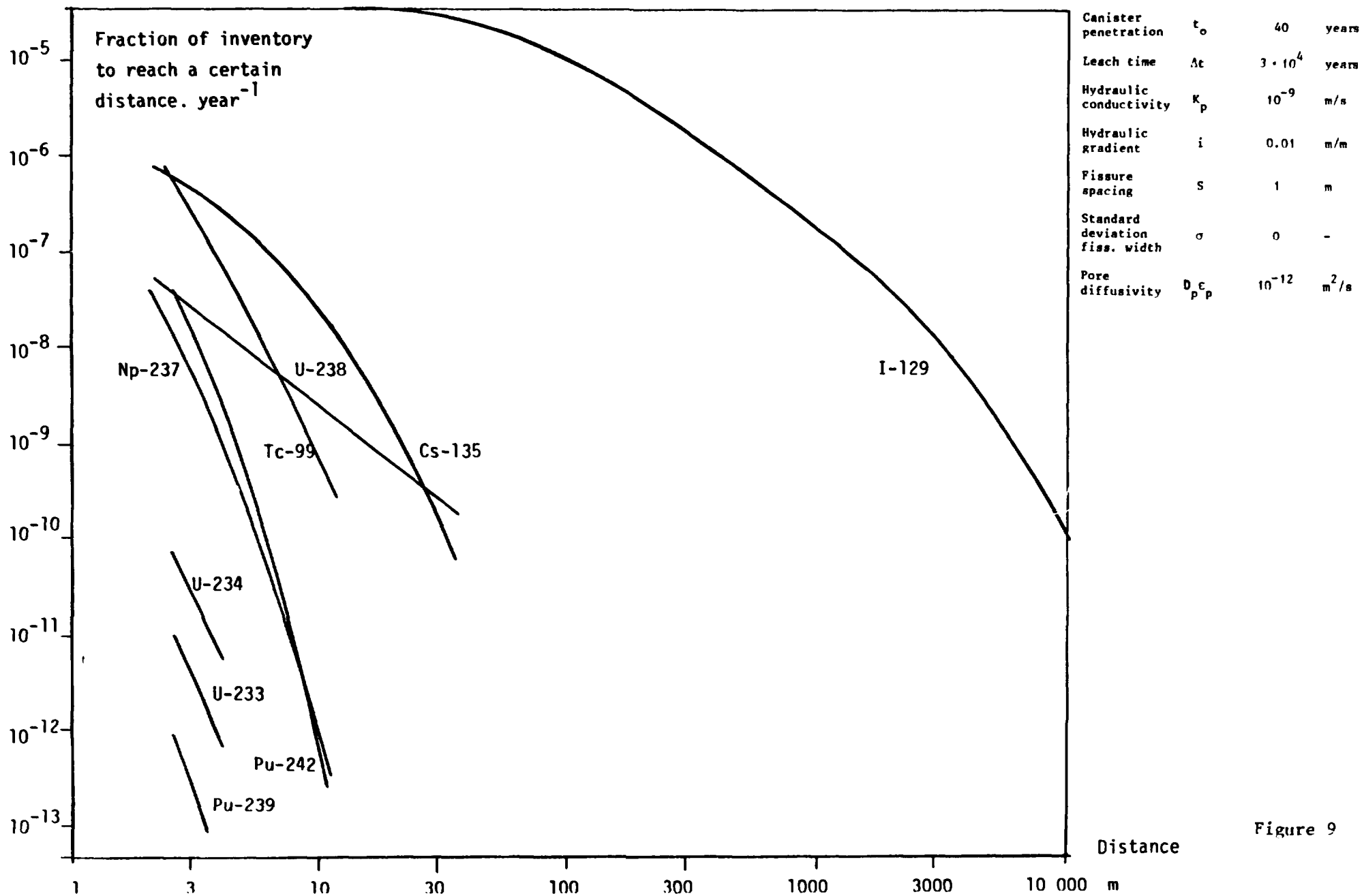


Figure 9

Fraction of inventory to reach a certain distance. year<sup>-1</sup>

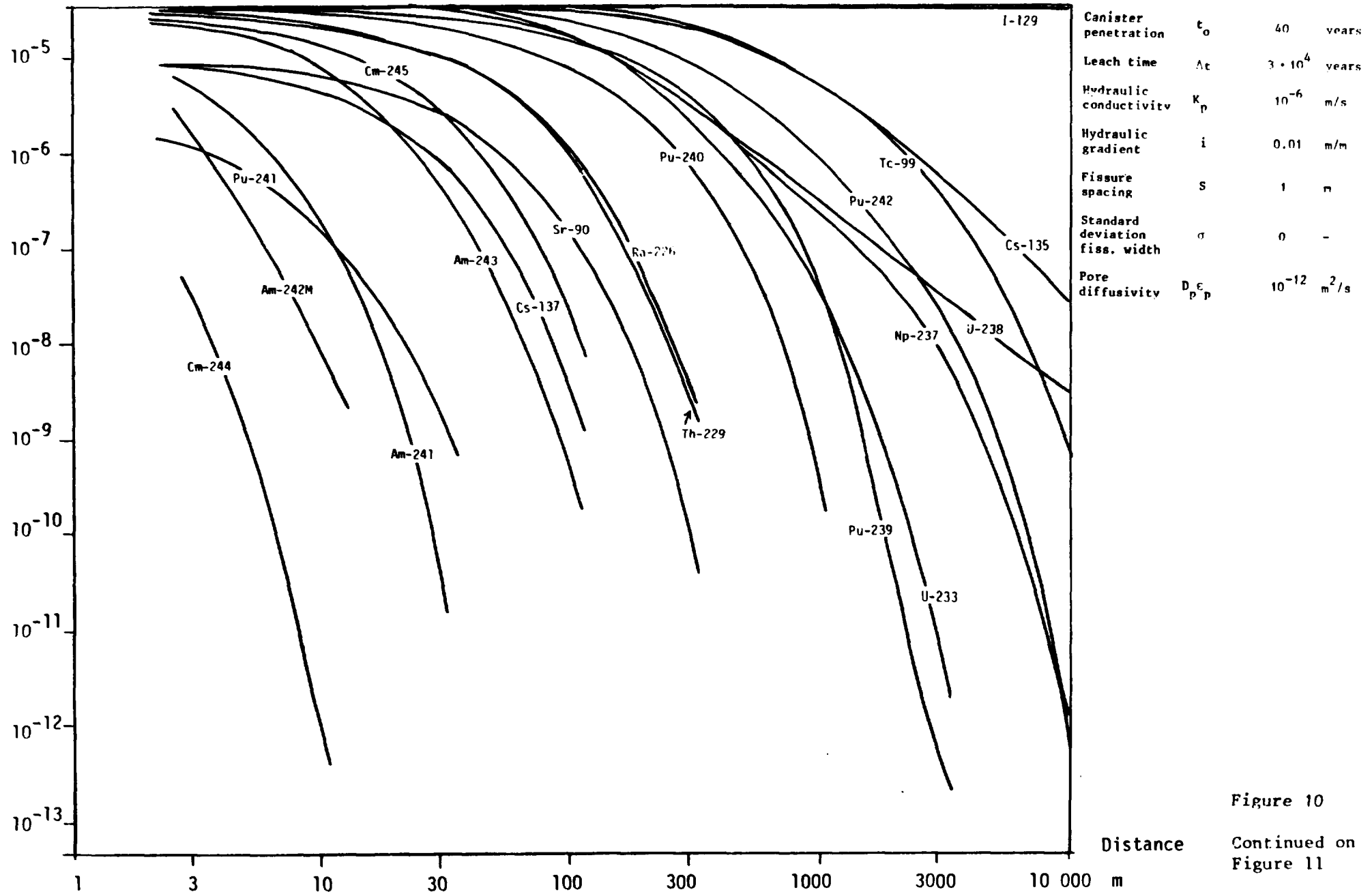


Figure 10

Continued on Figure 11

Fraction of inventory to reach a certain distance. year<sup>-1</sup>

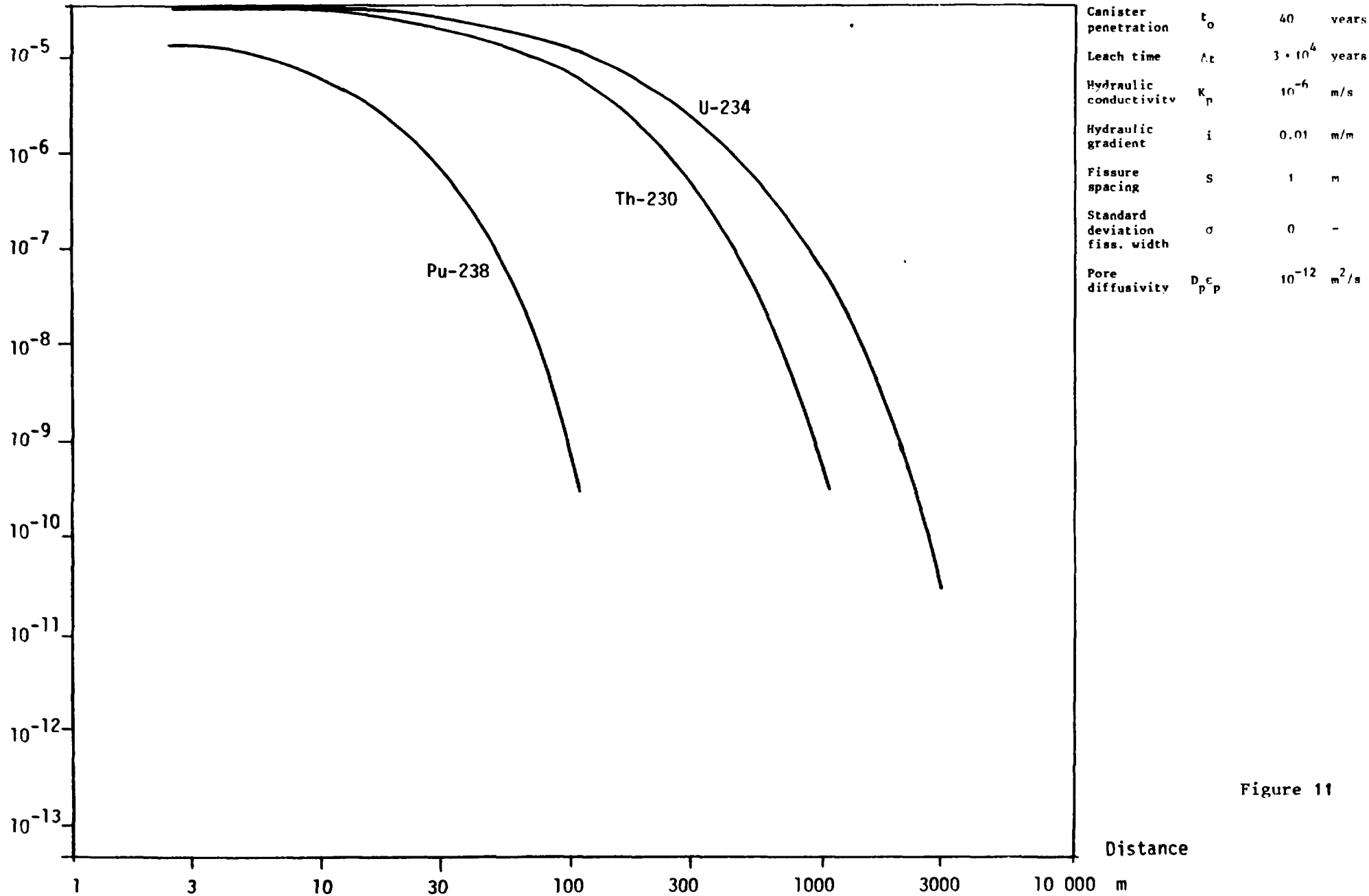


Figure 11

Distance

Fraction of inventory to reach a certain distance. year<sup>-1</sup>

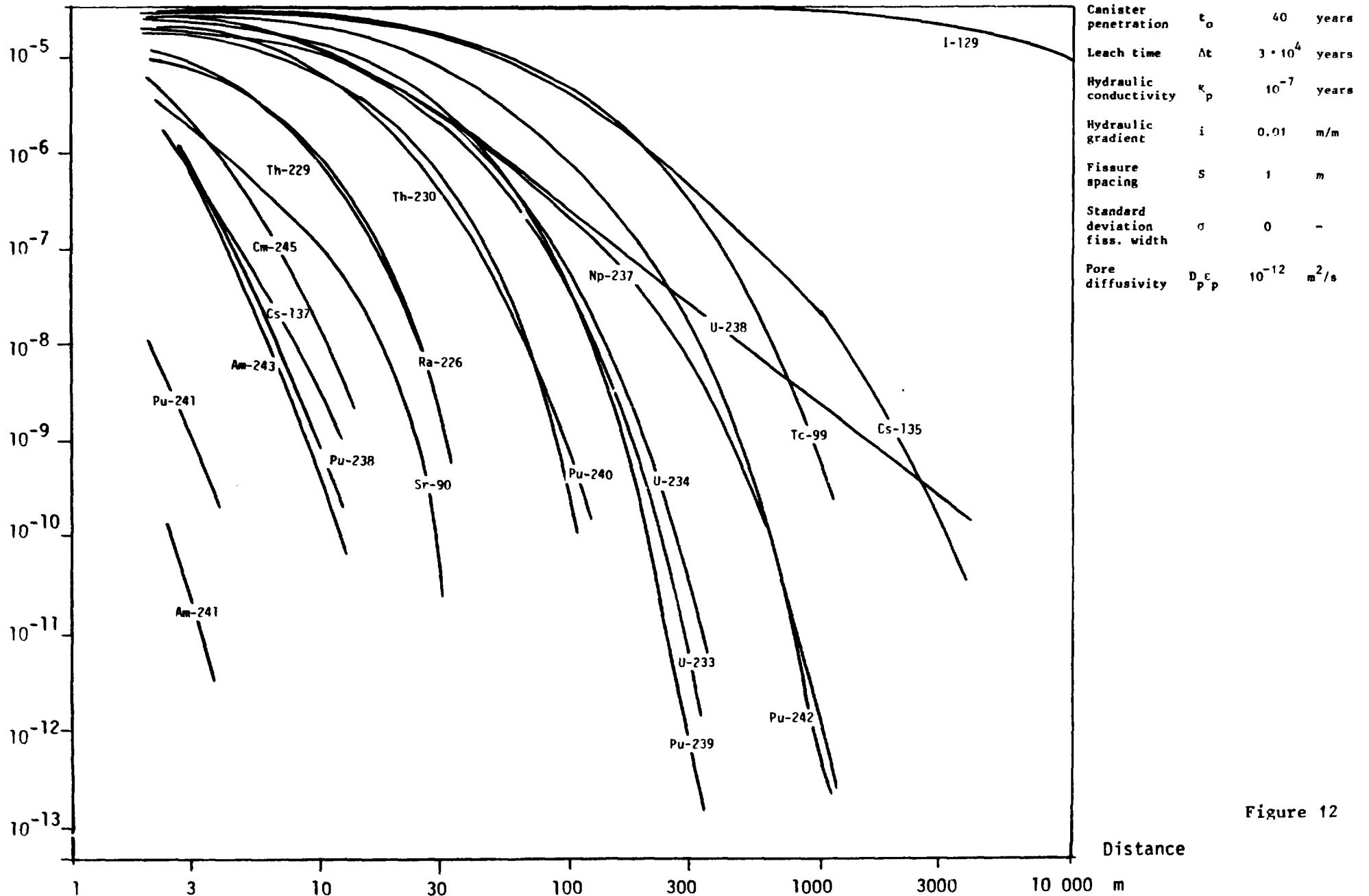


Figure 12

Fraction of inventory to reach a certain distance. year<sup>-1</sup>

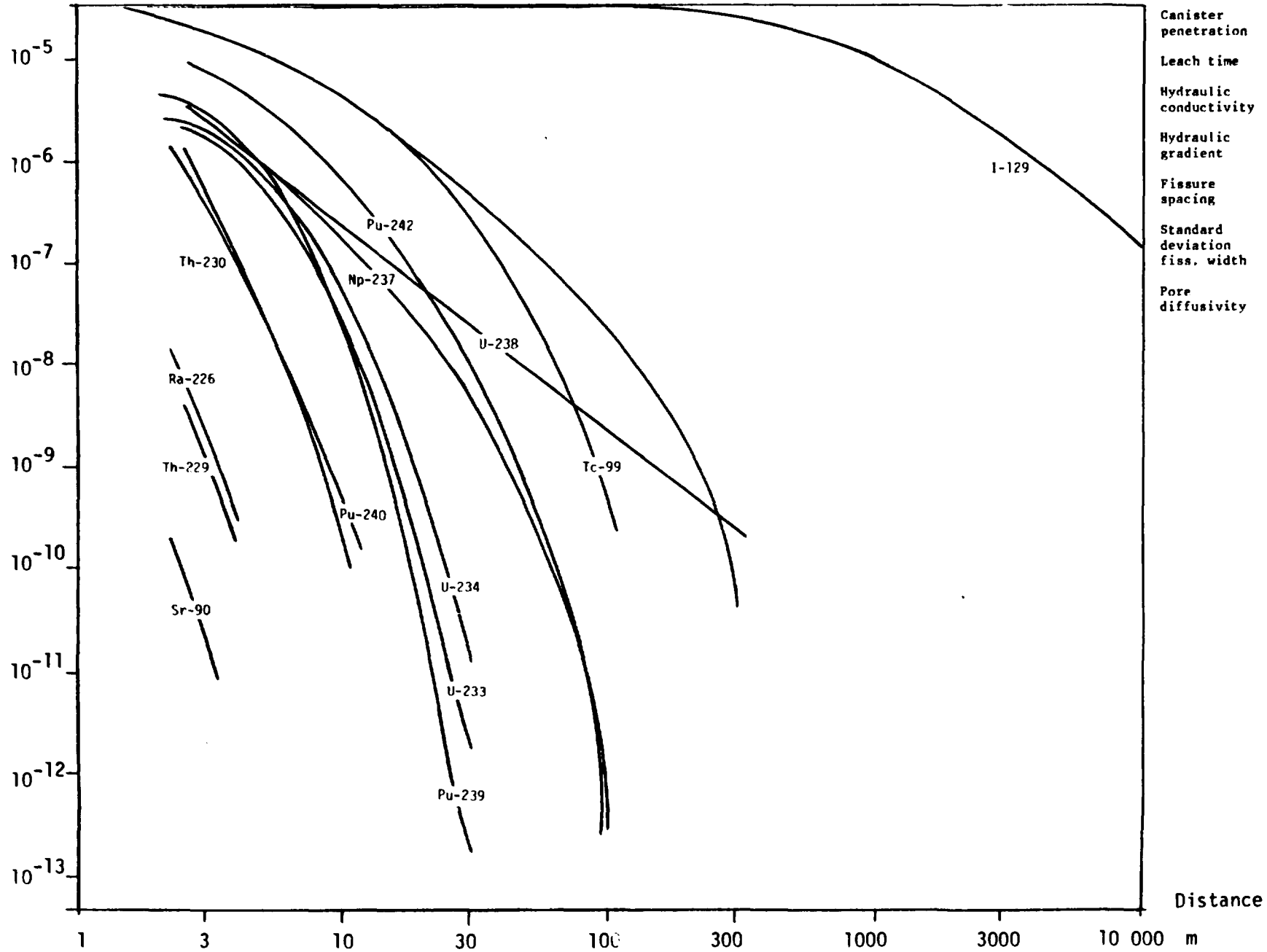
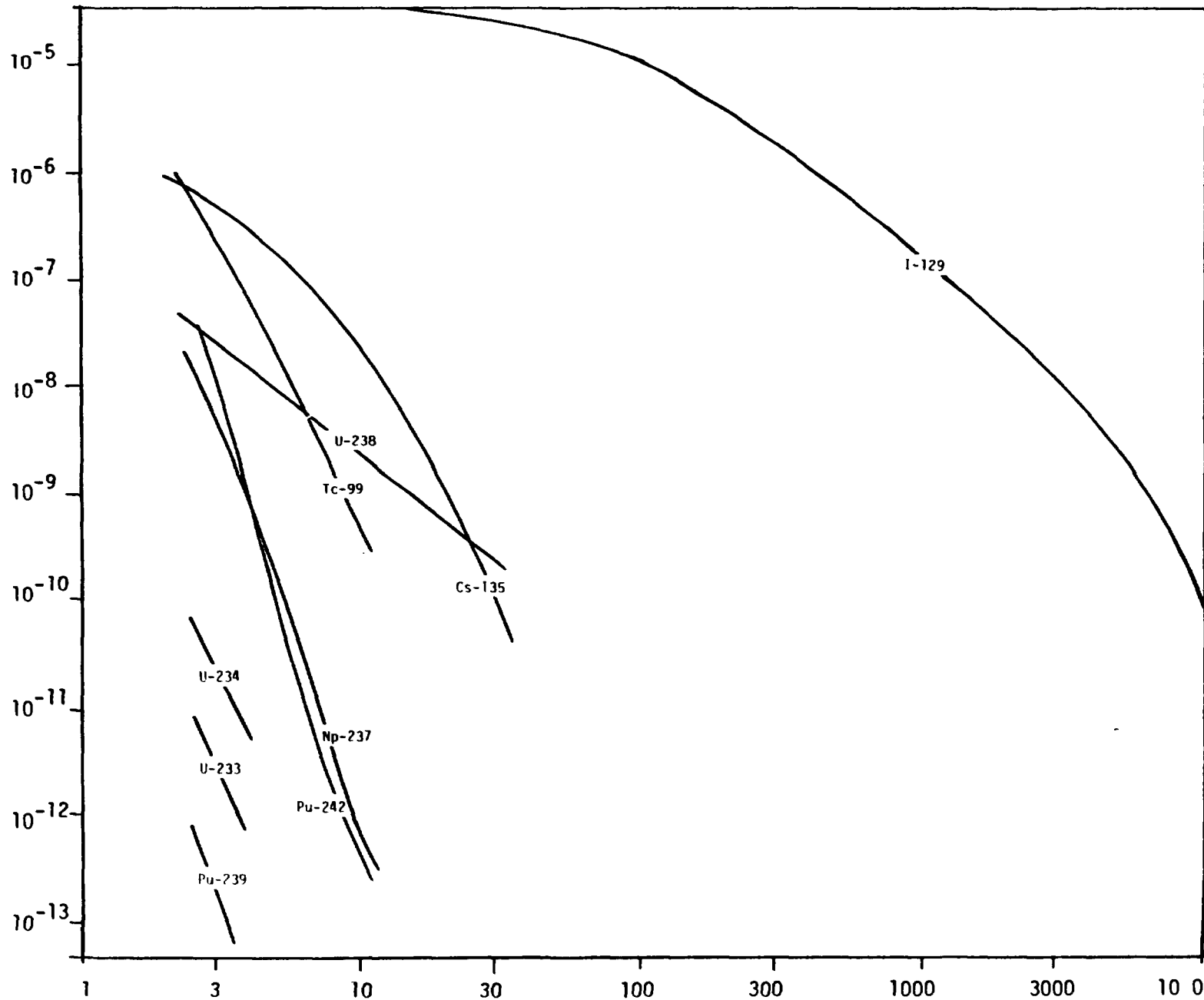


Figure 13



Fraction of inventory to reach a certain distance . year<sup>-1</sup>



Canister penetration	$t_o$	5000	years
Leach time	$\Delta t$	$3 \cdot 10^4$	years
Hydraulic conductivity	$K_p$	$10^{-9}$	m/s
Hydraulic gradient	$i$	0.01	m/m
Fracture spacing	$S$	1	m
Standard deviation fiss. width	$\sigma$	0	-
Pore diffusivity	$D_{pp}$	$10^{-12}$	m <sup>2</sup> /s

Figure 14

Distance

Fraction of inventory to reach a certain distance . year<sup>-1</sup>

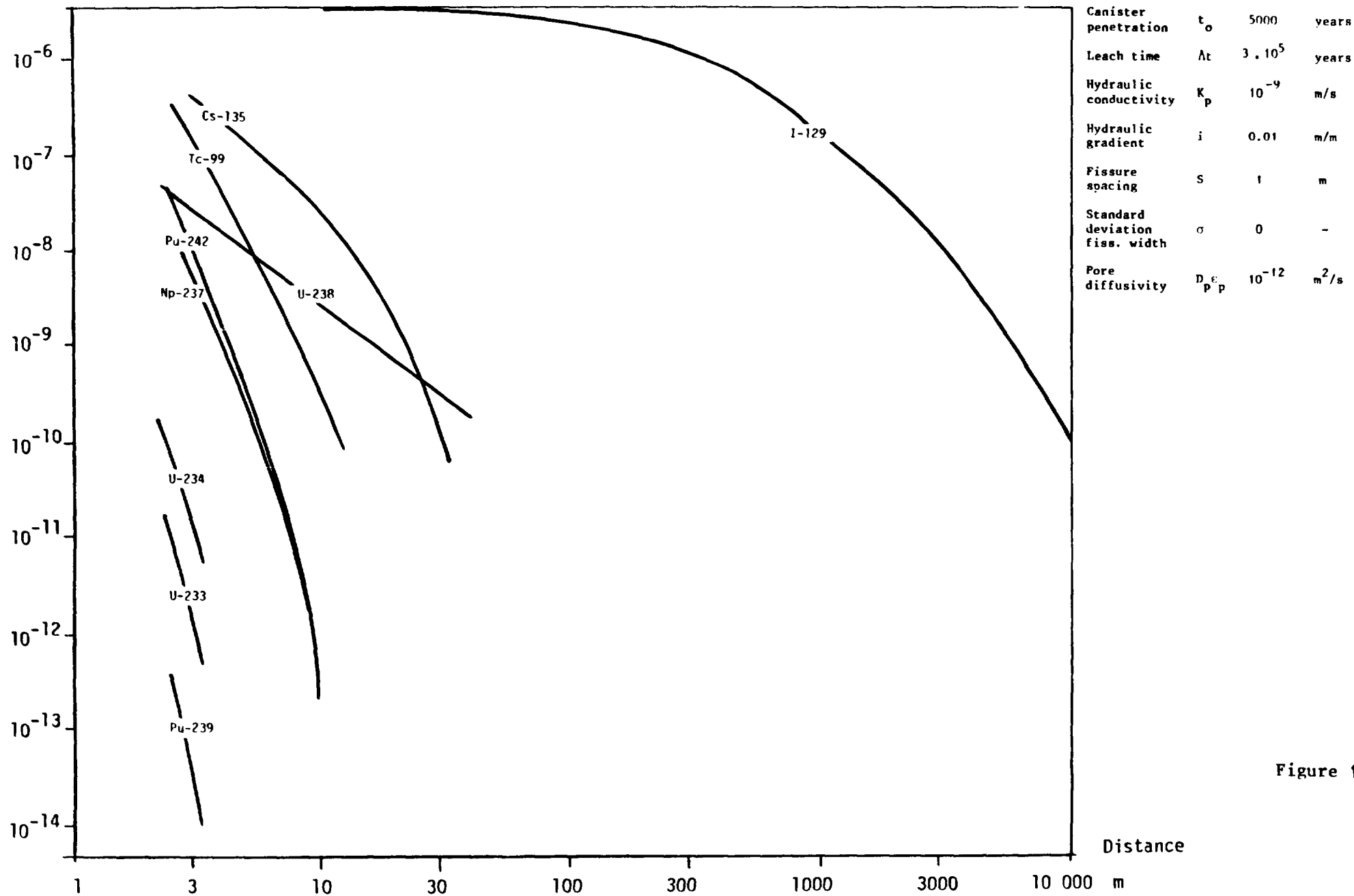


Figure 15

Fraction of inventory to reach a certain distance. year<sup>-1</sup>

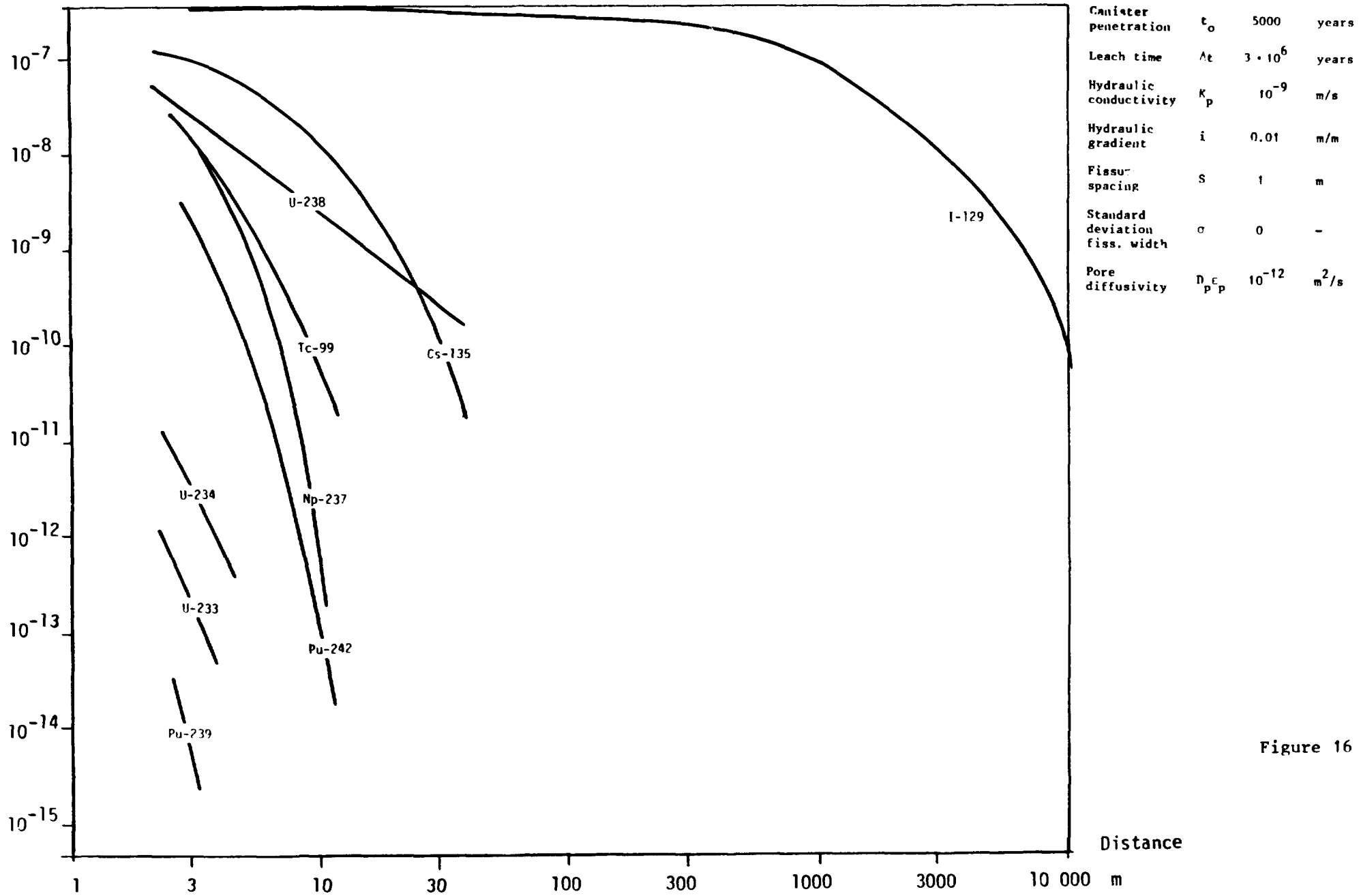
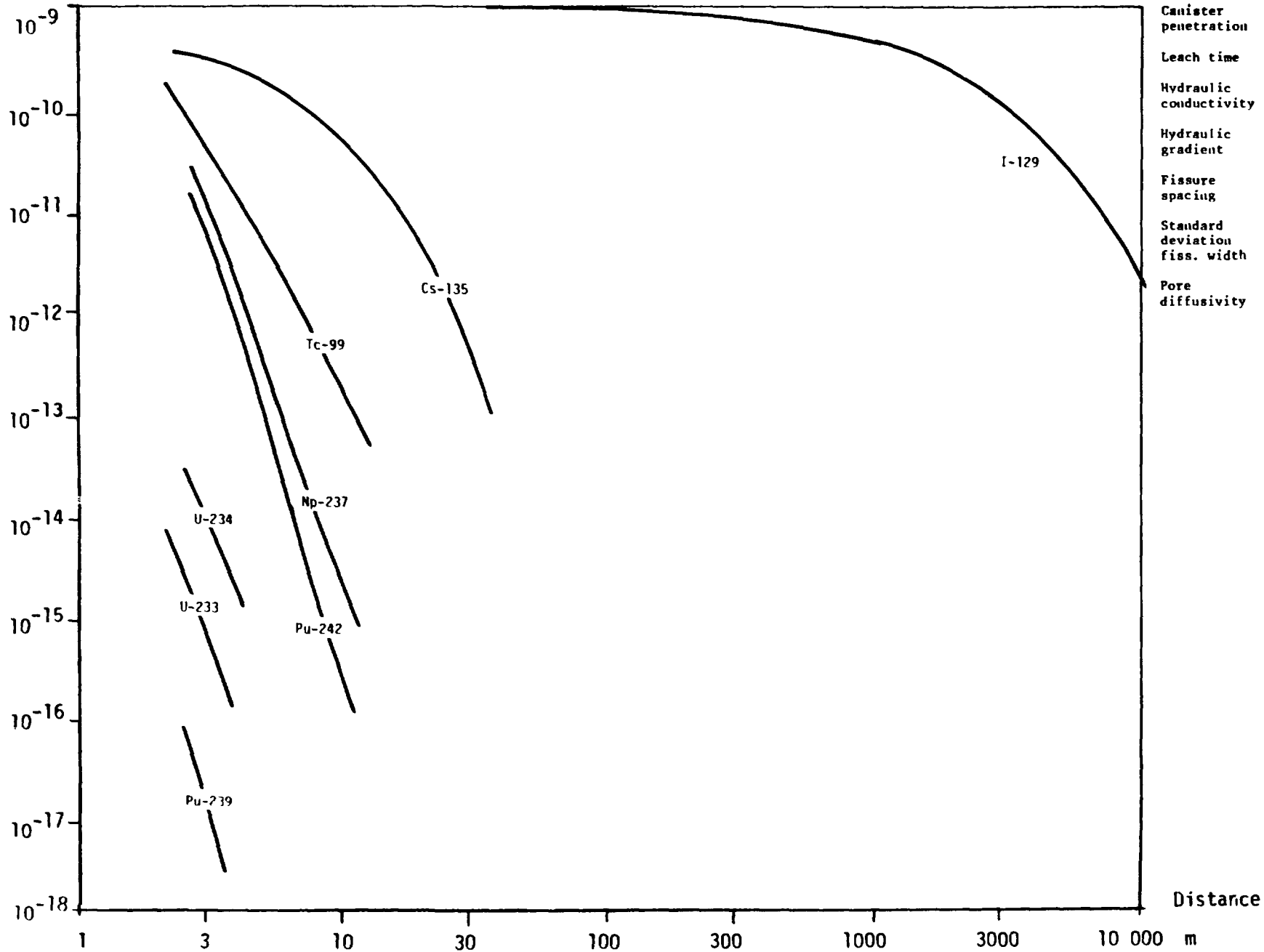


Figure 16

Fraction of inventory to reach a certain distance . year<sup>-1</sup>



Canister penetration	$t_0$	40	years
Leach time	$\Delta t$	$10^9$	years
Hydraulic conductivity	$K_p$	$10^{-9}$	m/s
Hydraulic gradient	$i$	0.01	m/m
Fissure spacing	$S$	1	m
Standard deviation fiss. width	$\sigma$	0	-
Pore diffusivity	$D_p \epsilon_p$	$10^{-12}$	m <sup>2</sup> /s

Figure 17

Fraction of inventory to reach a certain distance, year<sup>-1</sup>

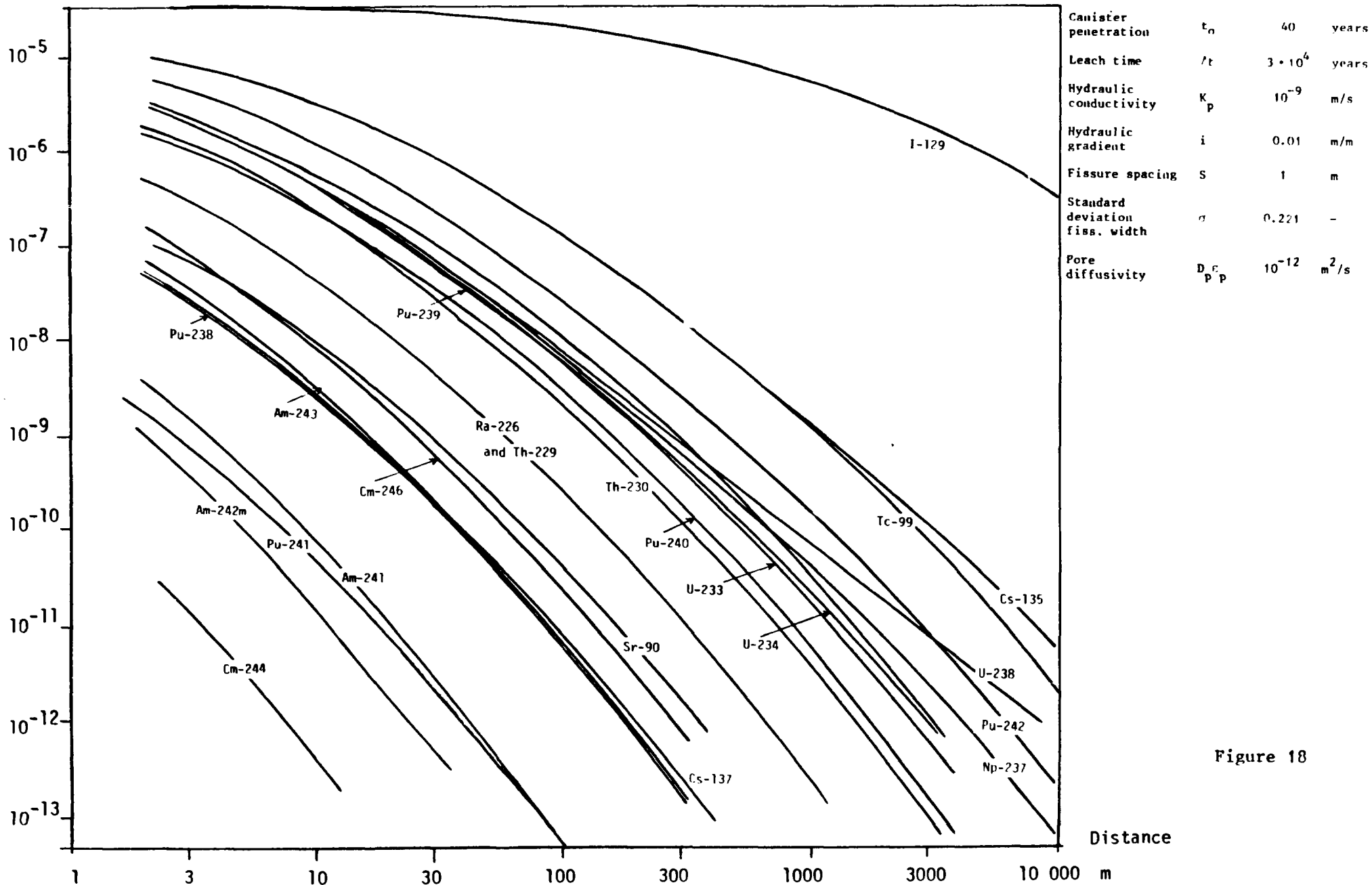


Figure 18

Fraction of inventory to reach a certain distance, year<sup>-1</sup>

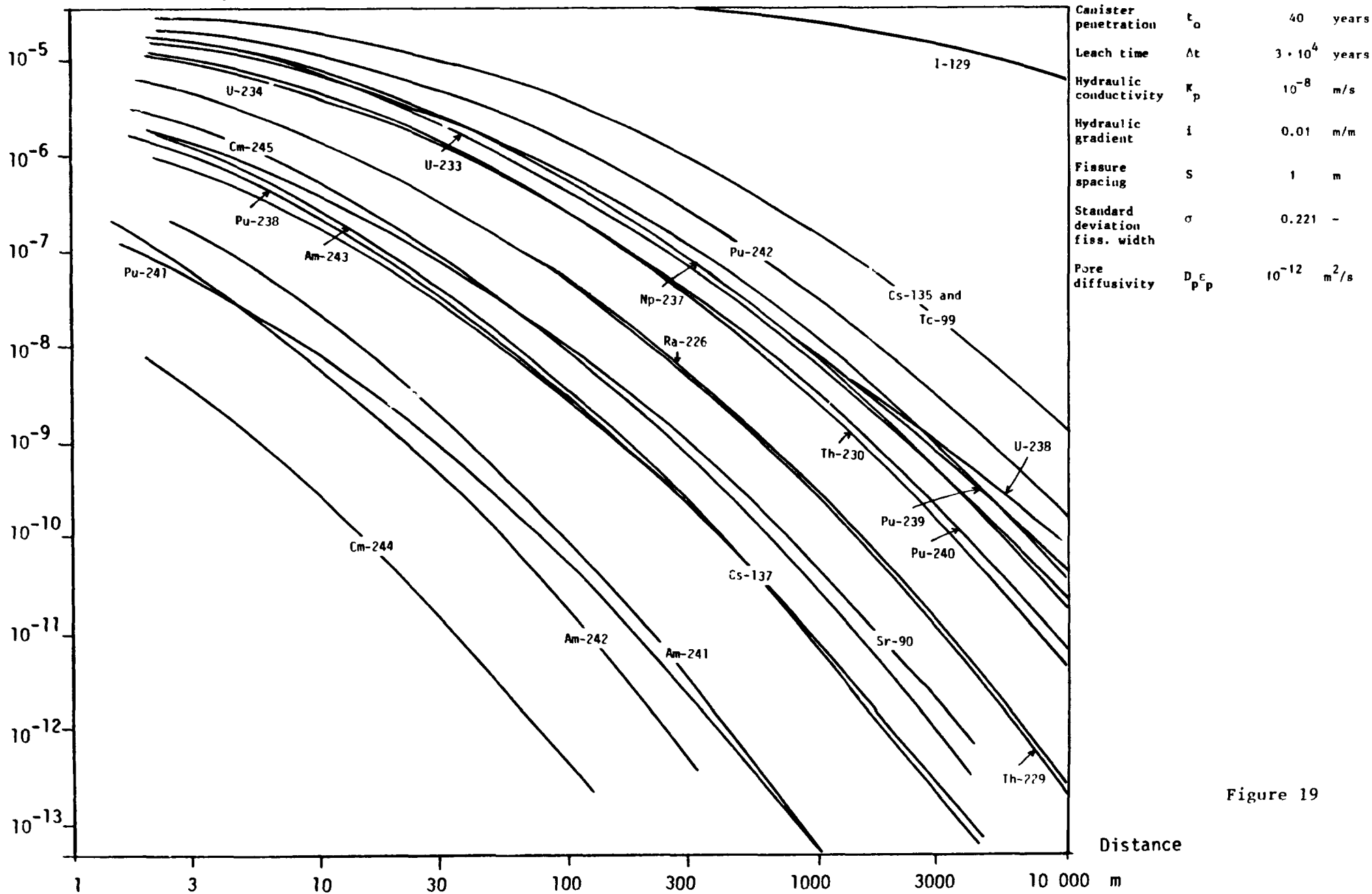


Figure 19

Fraction of inventory to reach a certain distance . year<sup>-1</sup>

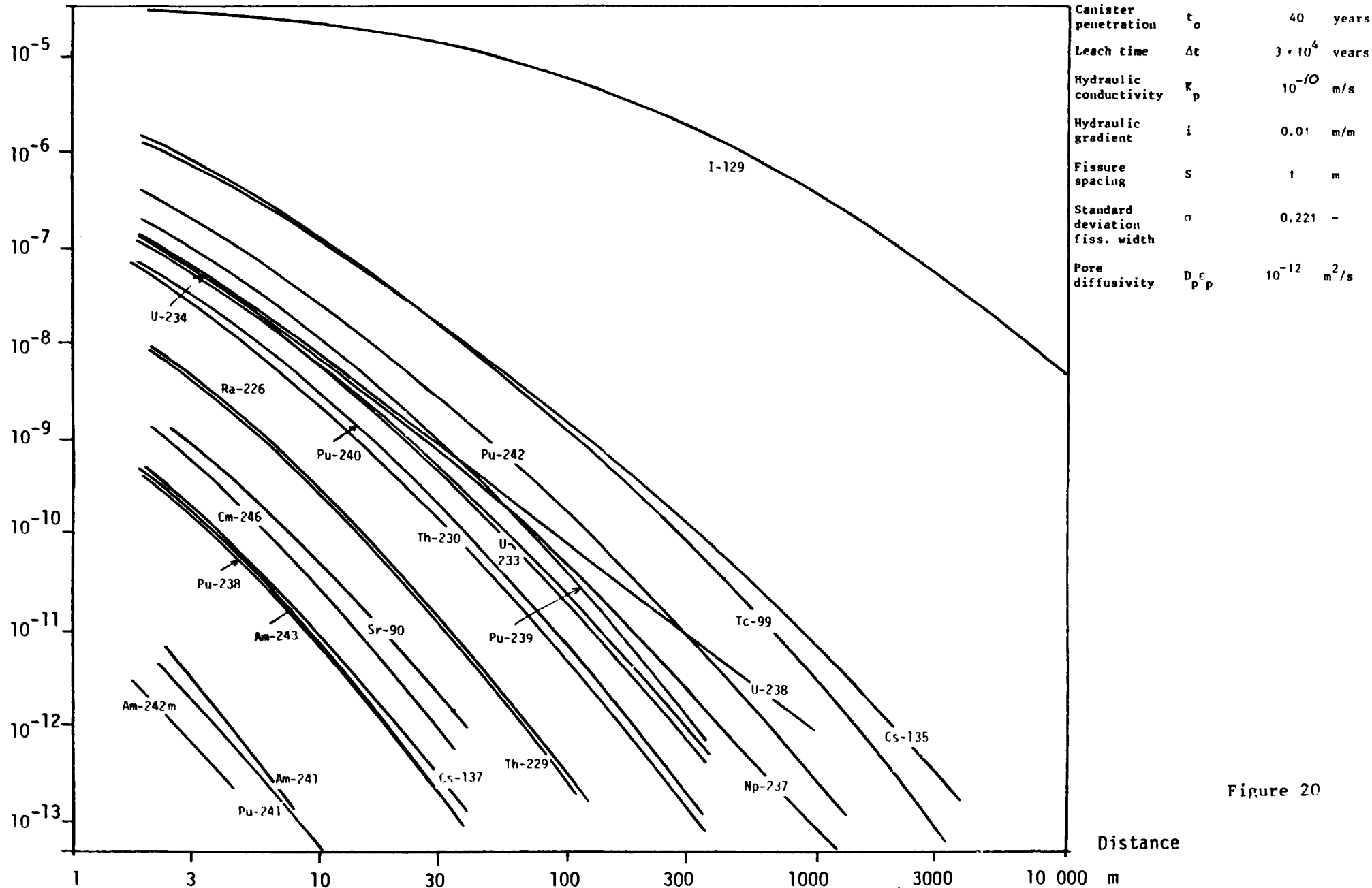


Figure 20

Fraction of inventory to reach a certain distance, year<sup>-1</sup>

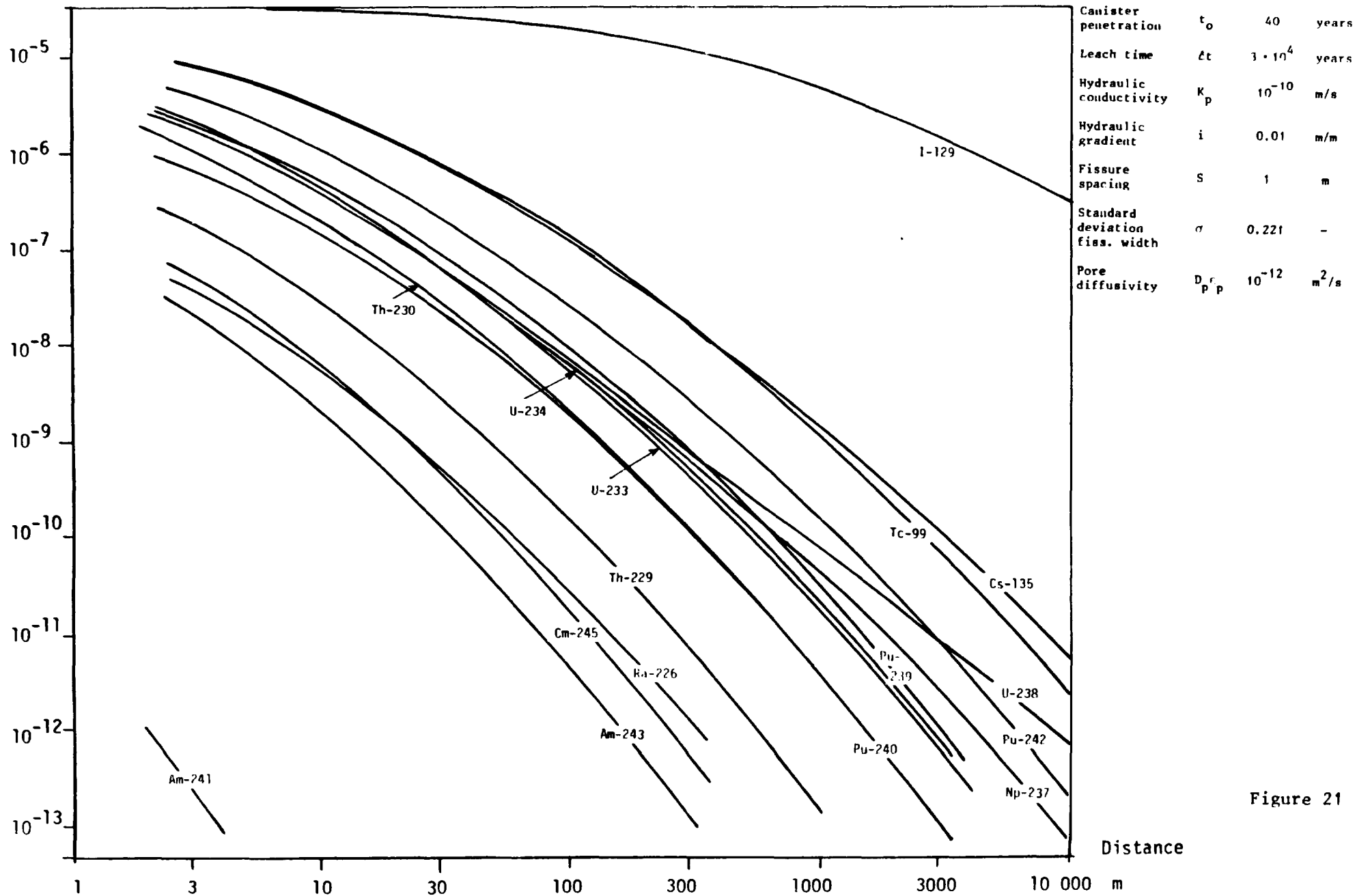


Figure 21



Fraction of inventory to reach a certain distance . year<sup>-1</sup>

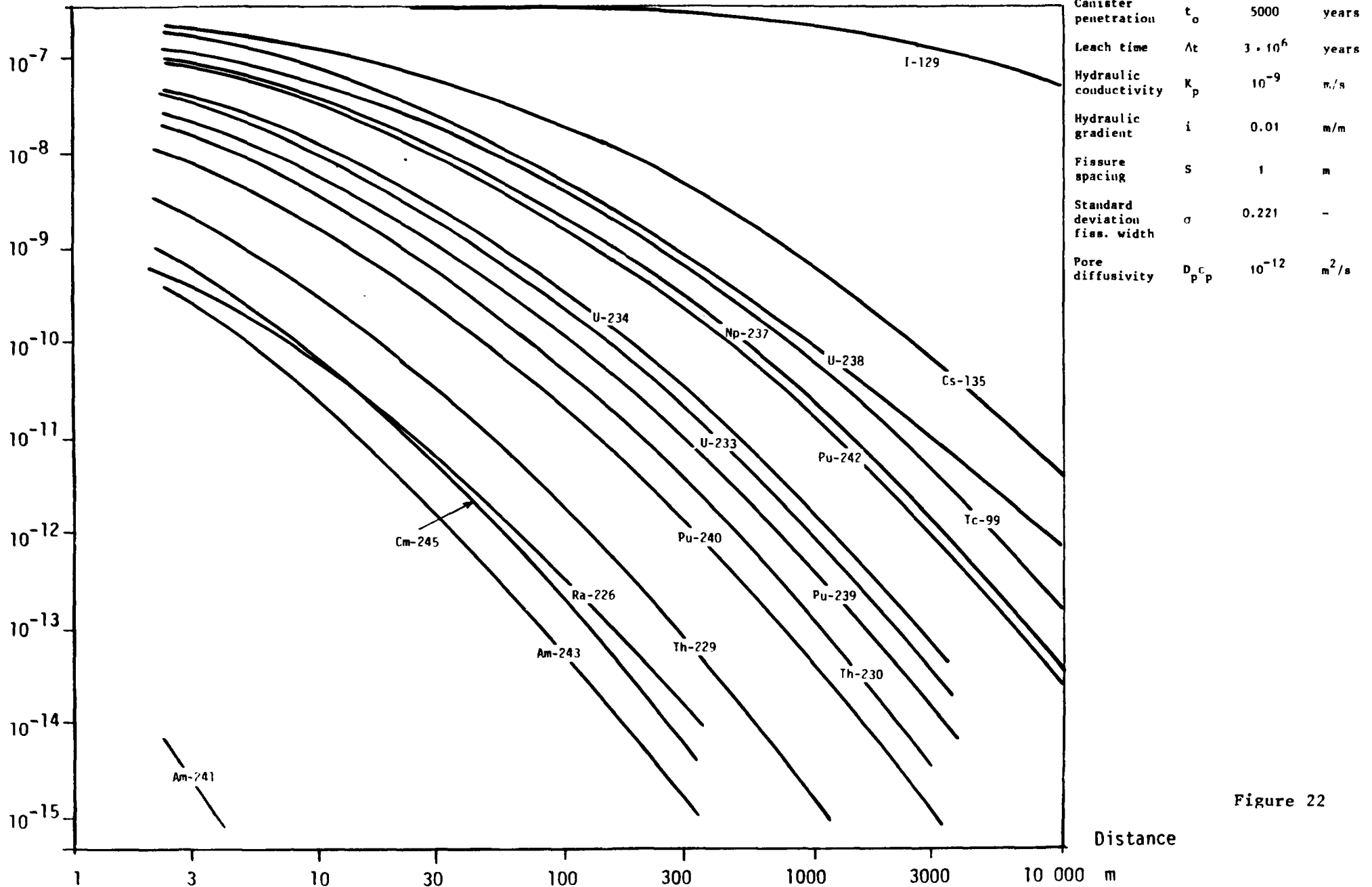


Figure 22

Fraction of inventory to reach a certain distance . year<sup>-1</sup>

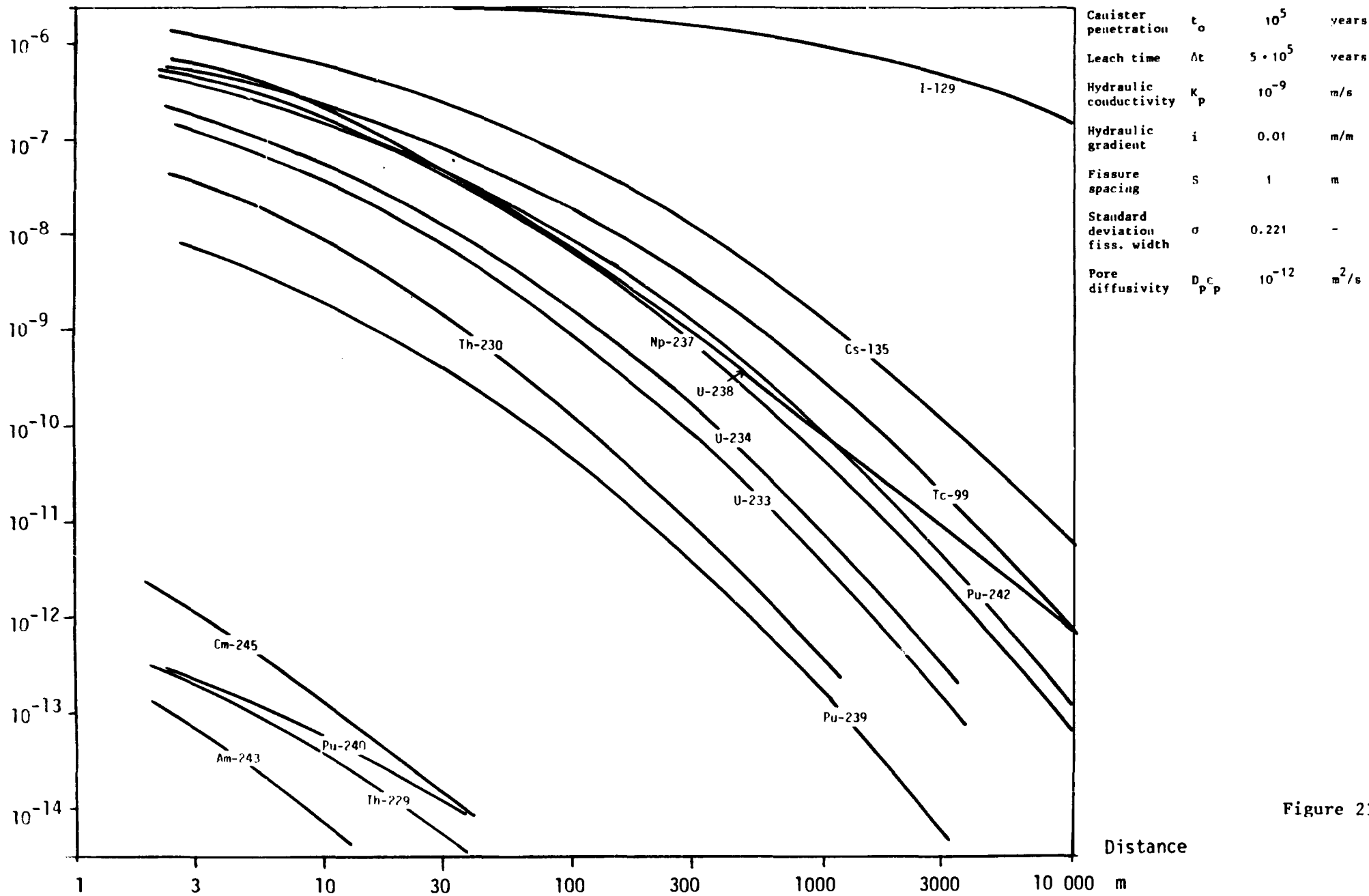


Figure 23

Fraction of inventory to reach a certain distance . year<sup>-1</sup>

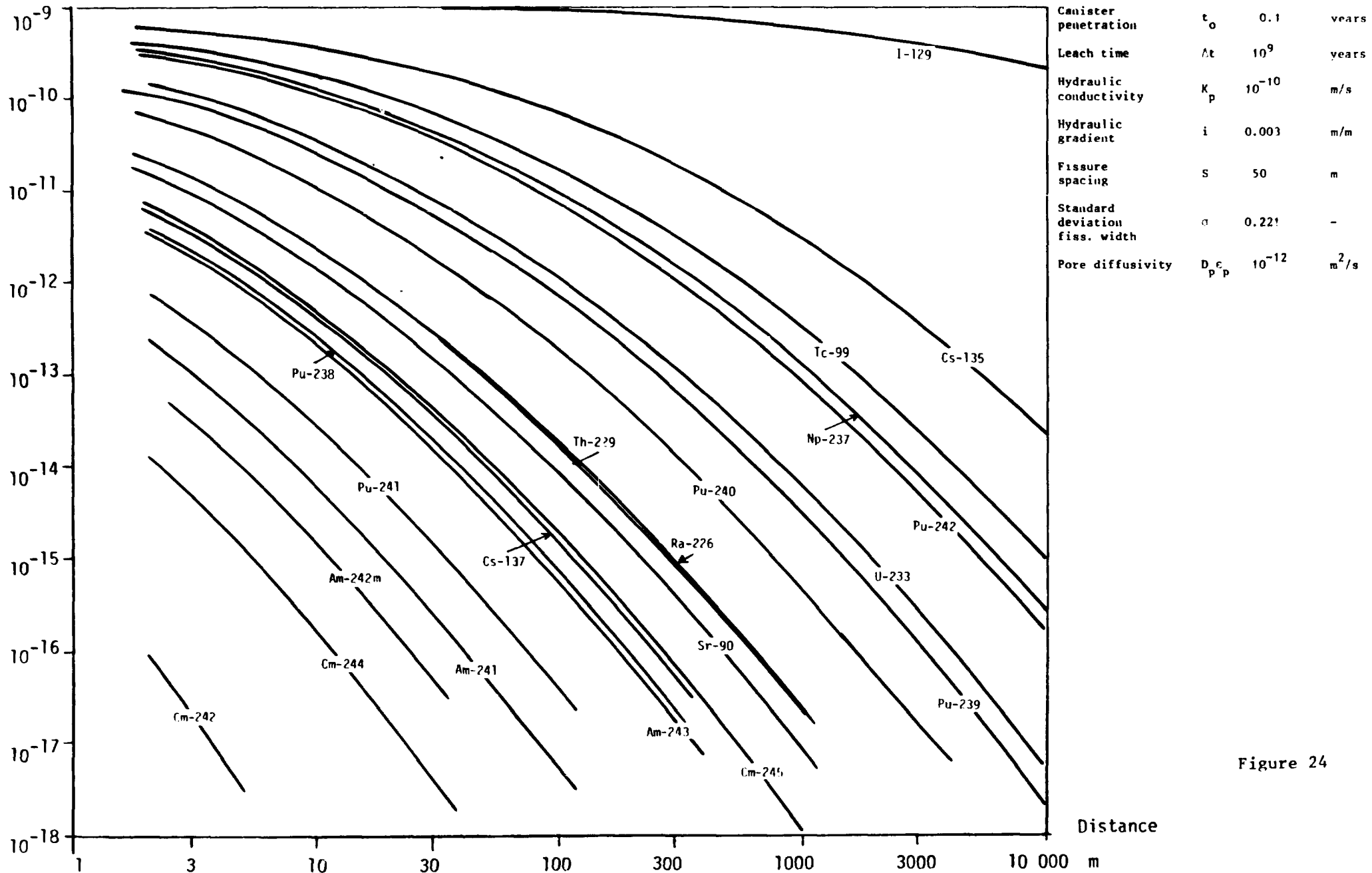


Figure 24

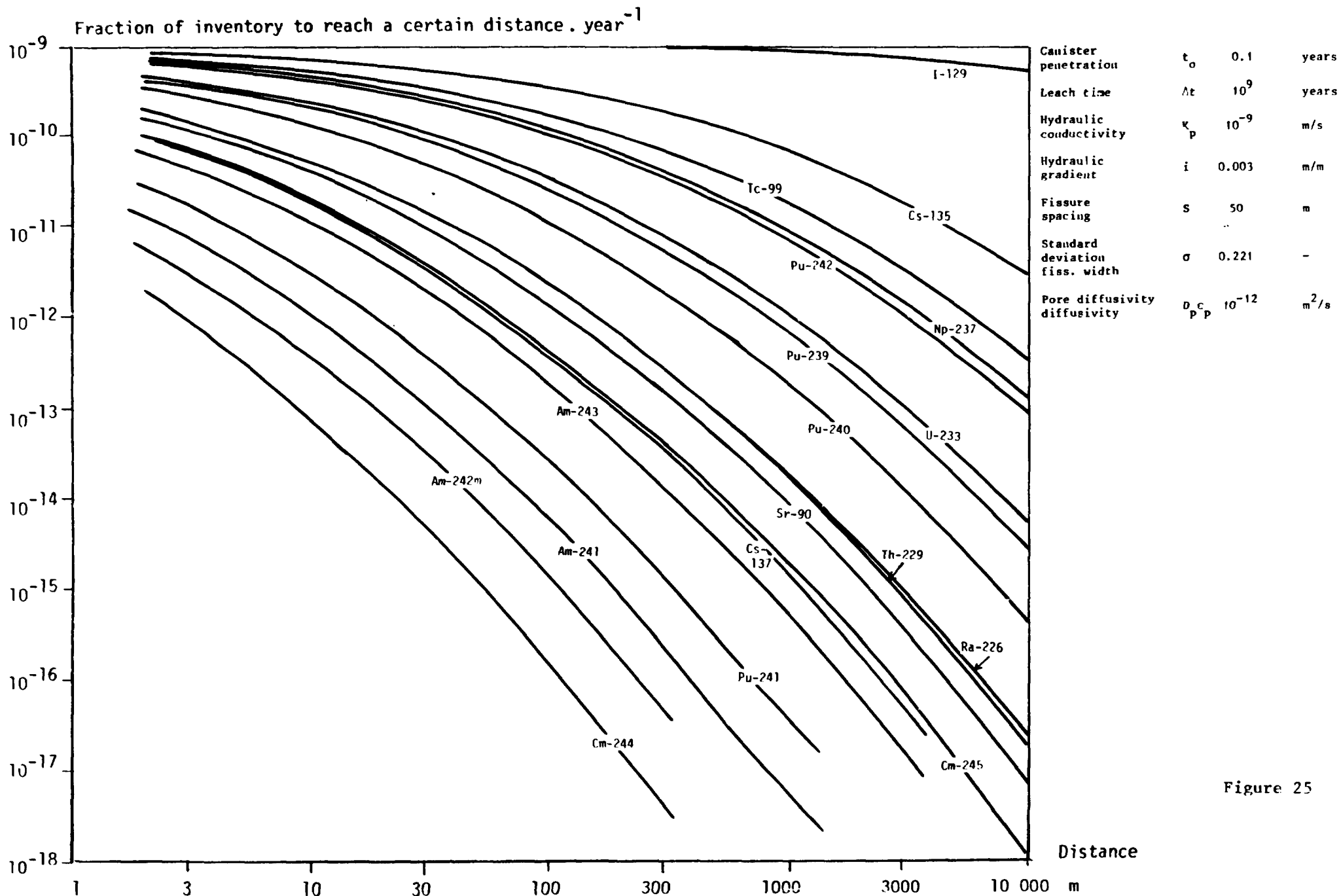


Figure 25

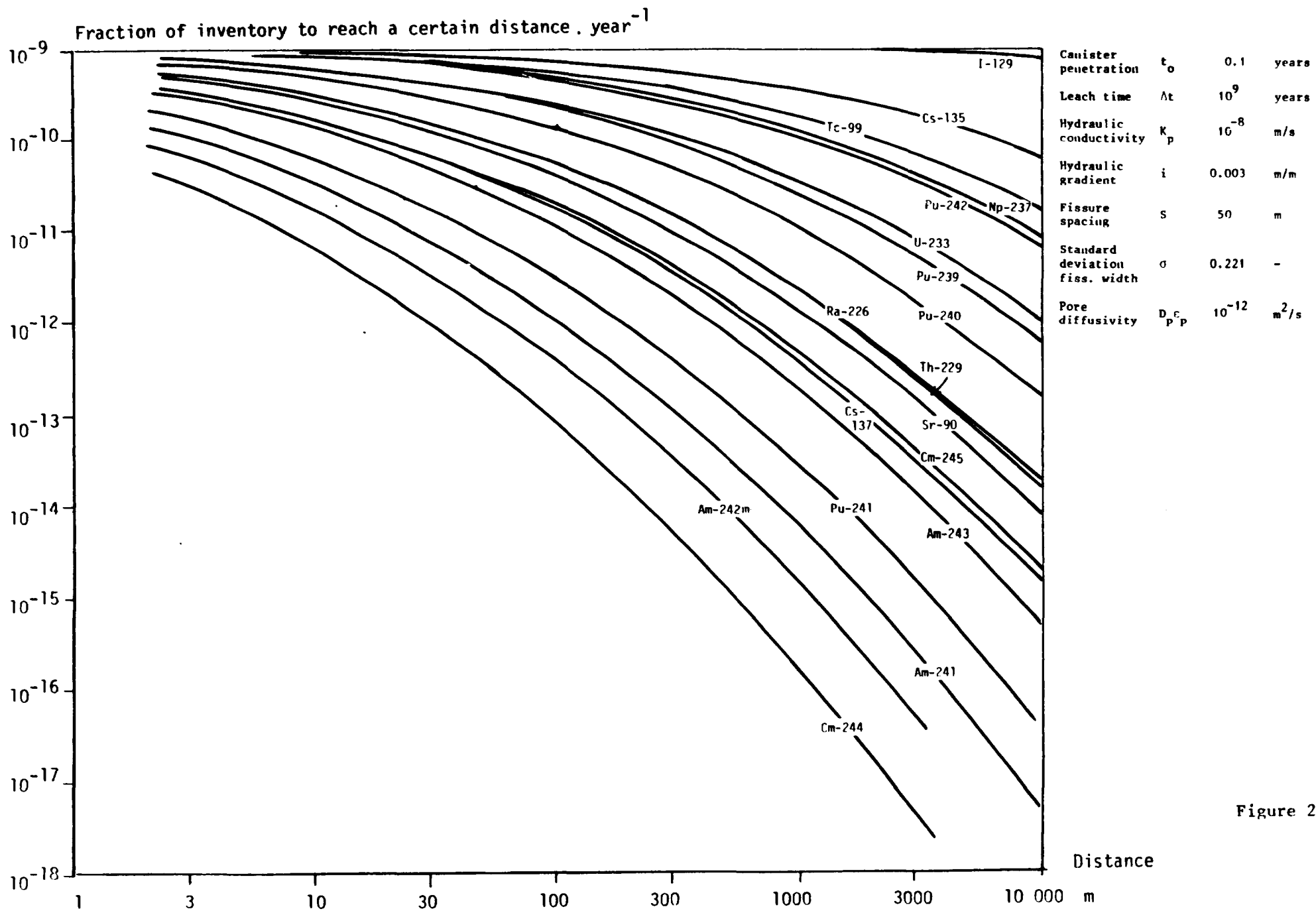


Figure 26

FÖRTECKNING ÖVER KBS TEKNISKA RAPPORTER

1977-78

TR 121 KBS Technical Reports 1 - 120.  
Summaries. Stockholm, May 1979.

1979

TR 79-28 The KBS Annual Report 1979.  
KBS Technical Reports 79-01--79-27.  
Summaries. Stockholm, March 1980.

1980

TR 80-26 The KBS Annual Report 1980.  
KBS Technical Reports 80-01--80-25.  
Summaries. Stockholm, March 1981.

1981

TR 81-17 The KBS Annual Report 1981.  
KBS Technical Reports 81-01--81-16  
Summaries. Stockholm, April 1982.

1982

TR 82-01 Hydrothermal conditions around a radioactive waste  
repository  
Part 3 - Numerical solutions for anisotropy  
Roger Thunvik  
Royal Institute of Technology, Stockholm, Sweden  
Carol Braester  
Institute of Technology, Haifa, Israel  
December 1981

TR 82-02 Radiolysis of groundwater from HLW stored in copper  
canisters  
Hilbert Christensen  
Erling Bjergbakke  
Studsvik Energiteknik AB, 1982-06-29

- TR 82-03 Migration of radionuclides in fissured rock:  
Some calculated results obtained from a model based  
on the concept of stratified flow and matrix  
diffusion  
Ivars Neretnieks  
Royal Institute of Technology  
Department of Chemical Engineering  
Stockholm, Sweden, October 1981
- TR 82-04 Radionuclide chain migration in fissured rock -  
The influence of matrix diffusion  
Anders Rasmuson \*  
Akke Bengtsson \*\*  
Bertil Grundfelt \*\*  
Ivars Neretnieks \*  
April, 1982
- \* Royal Institute of Technology  
Department of Chemical Engineering  
Stockholm, Sweden
- \*\* KEMAKTA Consultant Company  
Stockholm, Sweden
- TR 82-05 Migration of radionuclides in fissured rock -  
Results obtained from a model based on the concepts  
of hydrodynamic dispersion and matrix diffusion  
Anders Rasmuson  
Ivars Neretnieks  
Royal Institute of Technology  
Department of Chemical Engineering  
Stockholm, Sweden, May 1982
- TR 82-06 Numerical simulation of double packer tests  
Calculation of rock permeability  
Carol Braester  
Israel Institute of Technology, Haifa, Israel  
Roger Thunvik  
Royal Institute of Technology  
Stockholm, Sweden, June 1982
- TR 82-07 Copper/bentonite interaction  
Roland Pusch  
Division Soil Mechanics, University of Luleå  
Luleå, Sweden, 1982-06-30
- TR 82-08 Diffusion in the matrix of granitic rock  
Field test in the Stripa mine  
Part 1  
Lars Birgersson  
Ivars Neretnieks  
Royal Institute of Technology  
Department of Chemical Engineering  
Stockholm, Sweden, July 1982

