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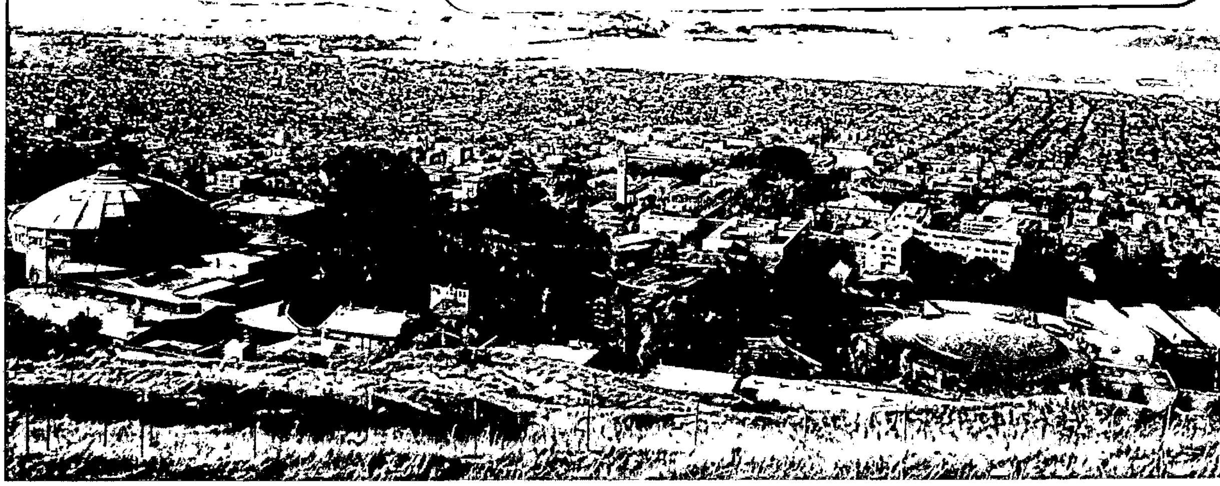

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HEAT-PIPE EFFECT ON THE TRANSPORT OF GASEOUS RADIONUCLIDES RELEASED FROM A NUCLEAR WASTE CONTAINER

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INTRODUCTION

When an unsaturated porous medium is subjected to a temperature gradient and the temperature is sufficiently high, vadose water is heated and vaporizes. Vapor flows under its pressure gradient towards colder regions where it condenses. Vaporization and condensation produce a liquid saturation gradient, creating a capillary pressure gradient inside the porous medium. Condensate flows towards the hot end under the influence of a capillary pressure gradient. This is a heat pipe in an unsaturated porous medium.[1–6]

We study analytically the transport of gaseous species released from a spent-fuel waste package, as affected by a time-dependent heat pipe in an unsaturated rock. For parameter values typical of a potential repository in partially saturated fractured tuff at Yucca Mountain, we found that a heat pipe develops shortly after waste is buried, and the heat-pipe's spatial extent is time-dependent. Water vapor movements produced by the heat pipe can significantly affect the migration of gaseous radionuclides.

ANALYSIS

To determine the vapor mass flux and the spatial and temporal extent of a heat pipe, coupled heat and mass transfer analysis is used. We consider a waste sphere of radius r_0 of surface area equal to that of actual waste containers, in an infinite homogenous porous medium. When a heat pipe develops, three zones can be identified, see Figure 1. Near the hot waste there is a vapor-only region $r_0 \leq r \leq r_1(t)$ where pore space is filled mostly by vapor. The intermediate zone $r_1(t) \leq r \leq r_2(t)$ is the heat pipe. Outside the heat pipe is a region where most of the pores are filled by liquid—the liquid zone $r_2(t) \leq r \leq \infty$.

Because decay heat decreases gradually with time, thermal equilibrium is maintained, and a quasi-steady-state analysis can be used. Furthermore, we use the "effective continuum" porous medium approach,[7] instead of treating fractures and matrix discretely. The ideal gas law is used for vapor. We assume fluid properties are constant. Gravitational effect is neglected. Non-condensible gases are not considered in this analysis.

Within a control volume, the flow of liquid water and water vapor is described by a continuity equation

$$\dot{m}_v + \dot{m}_\ell = 0 \tag{1}$$

where $\dot{m}$ is the mass flux and subscripts "v" and "ℓ" refer to vapor and liquid respectively. Vapor and liquid fluxes are given by Darcy's law

$$\dot{m}_v = -\frac{k_v(S)}{\mu_v}\frac{dp_v}{dr} \quad \text{and} \quad \dot{m}_\ell = -\frac{k_\ell(S)}{\mu_l}\frac{dp_\ell}{dr} \tag{2}$$

where p is the pressure, k is the permeability, μ is the dynamic viscosity and S is scaled liquid saturation. Scaled saturation is given by

$$S = (S_\ell - S_{\ell,irr})/(1 - S_{\ell,irr}) \tag{3}$$

where S_ℓ is the actual liquid saturation and $S_{\ell,irr}$ is the irreducible saturation.
Capillary pressure $p_c(S)$ is given by a known characteristic function

$$p_v - p_\ell = p_c(S) \tag{4}$$

The energy transport is described as

$$\dot{q}'' = -\lambda(S)\frac{dT}{dr} + \dot{m}_v h_{fg} \tag{5}$$

where T is temperature, $\lambda(S)$ is the thermal conductivity depending on saturation, h_{fg} is the enthalpy of vaporization and $\dot{q}''$ is the time-dependent heat flux from the waste; and $\dot{q}''$ is the driving force for the heat pipe.

Temperature and capillary and vapor pressures have the following relationship, assuming thermodynamic equilibrium, from combining Kelvin's and Clapeyron's equations

$$\frac{T}{T_0} = \frac{1 + p_c(S)/\rho_\ell h_{fg}}{1 - (T_0 R_v/h_{fg}) \log(p_v/p_0)} \tag{6}$$

where ρ is density, R_v is the vapor constant, T_0 is the boiling temperature at p_0, and p_0 is the atmospheric pressure at the repository horizon.

We seek to determine $r_1(t)$ and $r_2(t)$, the spatial extent of the heat pipe, defined as

$$\text{at} \quad r = r_1(t): \quad S = S_1, \quad \text{and} \quad T = T(S_1) \tag{7}$$

$$\text{at} \quad r = r_2(t): \quad S = S_\infty, \quad T = T(S_\infty), \quad \text{and} \quad p_v = p_0 \tag{8}$$

where S_1 is the saturation in the vapor zone and S_∞ is the ambient saturation.

We summarize our major findings as (9) to (14) below. The mathematical details will be reported separately. By solving (1) through (8), most of the variables in the system can be expressed as functions of saturation, we first solve for saturation

$$\frac{1}{r} = \frac{1}{r_1(t)} + \frac{4\pi h_{fg}}{\nu_v \dot{Q}\phi(t)} \int_{S_1}^{S(r)} \frac{p_c'(u)du}{\alpha(u)k_{eff}(u)} \tag{9}$$

where ν is the kinematic viscosity, $p_c'(S)$ is the derivative of capillary pressure with respect to saturation, $\dot{Q}\phi(t)$ is the heat generation history.

The effective permeability for the two-phase counter-current flow $k_{eff}(S)$ is

$$\frac{1}{k_{eff}(S)} = \frac{1}{k_v(S)} + \frac{(\nu_\ell/\nu_v)}{k_\ell(S)} \tag{10}$$

In (9) $\alpha(S)$ is the ratio of the convective heat-flux to $\dot{q}''$

$$\frac{1}{\alpha(S)} = 1 + \frac{\lambda(S)\nu_v/h_{fg}R_v\rho_v k_{app}(S)}{h_{fg}/T_0R_v - \log(p_v(S)/p_0)} \tag{11}$$

where $k_{app}(S)$ is the apparent permeability for the heat pipe, describing general fluid mobility of two-phase counter-current flow when there is a temperature gradient

$$k_{app}(S) = \frac{k_v(S)k_\ell(S)}{k_\ell(S) + (\nu_\ell\rho_v/\nu_v\rho_\ell)k_v(S)} \tag{12}$$

The vapor pressure is

$$p_v(S) - p_0 = \int_{S_\infty}^{S} \frac{p_c'(u)}{1 + k_v(u)\nu_\ell/k_\ell(u)\nu_v} du \tag{13}$$

In the vapor zone the primary heat transfer mode is conduction, thus the inner boundary of the heat-pipe zone $r_1(t)$ can be estimated as a result of quasi-steady-state heat conduction by

$$\frac{1}{r_1(t)} = \frac{1}{r_0} - \frac{T_w(t) - T(S_1)}{\dot{Q}\phi(t)/4\pi\lambda(S_1)} \tag{14}$$

where $T_w(t)$ is the waste-surface temperature, approximated from the solution of heat conduction in an infinite spherical medium with the heating rate $\dot{Q}\phi(t)$ at r_0 and with an initial temperature equal to ambient temperaure. $T(S_1)$ as well as $T(S)$ are obtained from (6), (7) and (13). Thus, $r_1(t)$ is known. Then, $r_2(t)$ is known through (8) and (9). The functions $T(S(r))$, $p_v(S(r))$ and $\alpha(S(r))$ are then obtained from (6) through (13).

Finally, the vapor mass flux is

$$\dot{m}_v(r,t) = \rho_v(r,t)v_v(r,t) = \frac{\alpha(S(r))\dot{Q}\phi(t)}{4\pi h_{fg}r^2} \tag{15}$$

where $\alpha(S(r))$ is obtained from (11) through (14).

When the "effective continuum" model[7] is applied to tuff with dry fractures, and $k_f \gg k_m$, we have

$$k_v(S) \approx k_f \qquad \text{and} \qquad k_\ell(S) \approx k_m k_{r\ell,m}(S) \tag{16}$$

where subscripts "f" and "m" are for fractures and matrix and "$r\ell$" is for relative permeability for liquid.

To determine the effect the heat pipe has on the transport of gaseous radionuclide, we solve the advective-diffusion equation for a gaseous species for the system with a heat pipe, and compare the species concentration with one calculated without a heat pipe. If D is the effective diffusion coefficient of the species in the gas phase and C is its concentration in the gas phase, the governing equation for transport is

$$\frac{1}{r^2}\frac{d}{dr}\left(-r^2 D\frac{dC}{dr}\right) + \frac{1}{r^2}\frac{d}{dr}(r^2 vC) = 0, \qquad r_0 \le r < \infty. \tag{17}$$

where v is the vapor velocity obtained from (15). Here we have assumed that the amount of vapor is much larger than the amount of the species so that vapor acts like a carrying gas. We have neglected sorption and radioactive decay.

The boundary conditions are

$$C(r_0) = C_0 \qquad \text{and} \qquad C(\infty) = 0 \tag{18}$$

The solutions for (17) and (18) will be reported in detail separately.

NUMERICAL ILLUSTRATIONS

We use fluid properties from steam tables in standard texts.[8] In particular, we use the values in Table I. The waste sphere has a radius of 0.73 m. We assume that the repository will be in the Topopah Spring welded tuff unit.[9] Most of the matrix and fracture properties are from experimental results of Peters *et al.*[10], including a characteristic curve fitted by the van Genuchten method.[11] The ambient tuff temperature is assumed to be 25°C.[9] We consider heating from a spent-fuel waste package containing ten-year-old waste, with the following heating history

$$\phi(t) = 0.7707e^{-0.02689t} + 0.1932e^{-0.0022t} + 0.02163e^{-0.00005343t}, \qquad 0 \le t \le 50,000 \tag{19}$$

Other properties are taken from a simulation study by Tsang and Pruess.[12]

The shaded area in Figure 2 shows the extent of the heat pipe as a function of time. We also show the waste surface temperature and normalized waste heat generation, $\phi(t)$. The extent of the heat pipe is a function of the heating by the waste. When the tuff temperature is lower than the $T(S_\infty) = 96.18$°C, the heat pipe disappears. For the waste sphere we consider, the heat pipe exists from eight days after emplacement to 40 years.

Figure 3 shows the temperature and vapor pressure within the heat pipe as a function of saturation. They both decrease with saturation. The temperature gradient becomes small near the cold end (high saturation region), corresponding to the decrease of conductive heat flux. The vapor pressure gradient becomes great

Table I. Parameter values

Parameter	Units	Value	Source
Matrix permeability, k_m	[m²]	1.9×10^{-18}	[10]
Fracture permeability, k_f	[m²]	1.8×10^{-14}	[10]
Thermal conductivity, $\lambda(S_\ell)$	[W/m-°C]	$1.74 + 0.6 \times S_\ell$	[12]
Capilllary pressure, $p_c(S)$	[MPa]	$1.7265(S^{-2.253} - 1)^{0.5562}$	[10]
Characteristic curve, $k_{r\ell,m}(S)$		$\sqrt{S}[1-(1-S^{2.253})^{0.4438}]^2$	[10]
Ambient saturation, $S_{\ell,\infty}$		0.87	[12]
Irreducible saturation, $S_{\ell,irr}$		0.0801	[10]
Boiling temperature, T_0	[°C]	96.0	SCP,[9] 7-40
Spent-fuel waste package initial heating rate, $\dot{Q}$	[kW]	3	SCP,[9] 7-23
Atmospheric pressure, p_0	[MPa]	0.088	SCP,[9] 7-40

near the cold end, corresponding to the increase of convective heat flux. Temperatures at the two boundaries of the heat pipe are $T(S_1) = 132$°C for the hot end and $T(S_\infty) = 96.18$°C (because of the capillary effect, this value is greater than the boiling point, which is 96°C at repository horizon) for the cold end.

Figure 4 shows liquid saturation and vapor velocity at different times. Vapor velocity increases with radial distance due to vaporization along the heat pipe. Vapor flows towards the cold end. The temperature at $r > r_2(t)$ is below 96.18°C, and all the vapor reaching $r > r_2(t)$ condenses, dropping the vapor velocity to zero. Due to the variation of the heat-pipe zone, the magnitudes of vapor velocity also vary. The maximum velocity at each time happens at the cold end. The maxima are high when the heat source is strong and the heat pipe is close to the waste. They are lower when the heat source is weaker and the heat pipe is farther from the waste. Because fracture permeability is much larger than matrix permeability, and the fracture is dry, vapor flow is mainly in fractures. Our results of vapor velocity are similar to numerical simulation obtained by other workers.[7]

Figure 5 shows the mass flux of a gaseous radionuclide at the waste surface $\dot{m}_0$ normalized by its concentration $C_0(t)$, also at the waste surface, as a function of time. A diffusion coefficient of 30 m²/a is used. The dotted line is the maximum vapor velocity, v_{max}. The maximum v_{max} is 5000 m/a, at eight days after emplacement. This velocity is about 1000 times the gas velocity if there is no heat pipe. Thus, the radionuclide flux is higher with a heat pipe than without one, especially at early time. At later time this effect becomes smaller, but this enhanced radionuclide flux returns again towards the end of the heat-pipe's existence. Comparing with the diffusion-only case, the presence of a heat pipe increases the release rate of the radionuclide by 1.3 to 7 times due to increased vapor movement.

Figure 6 shows the concentration of a gaseous radionuclide as a function of radial distance, at three years after emplacement. Due to the variation in vapor velocity (shown as dotted line), the concentration becomes very small near the cold end of the heat pipe and then increases sharply, causing back diffusion in the heat pipe. Such details of the concentration profile would not be observed without a heat pipe analysis.

CONCLUSIONS

From this analysis, we found

(1) For the waste we analyzed, a heat pipe would exist from eight days to 40 years after emplacement. The heat pipe extends from the waste surface to about three meters from the center of the waste sphere.

(2) Water vapor velocity increases with radial distance within the heat pipe due to vaporization, and its magnitude varies with time because of the time-varying heat pipe. The maximum velocity is 1000-fold greater than the local air velocity if there were no heat pipe. This result agrees with previous results by other investigators.

(3) In the heat pipe, the concentration of the gaseous species in water vapor decreases near the hot end and increases near the cold end due to vapor movement.

(4) If the gaseous species release mechanism maintains a near-constant concentration of gaseous species in the short period of time in the gas outside and near the waste container surface, the mass flux of transport of that species would be increased 1.3 to 7 times greater than if there were no heat pipe. This implies, however, that if the release rate of gaseous species is affected little by the concentration of that species outside the container, the heat pipe can have little effect on the transport rate of that species.

Our analysis shows there is the possiblity of creating a heat pipe at the potential repository at Yucca Mountain, and that a heat pipe would have significant effect on the transport of gaseous radionuclides. Further studies of such a phenomenon appears warranted.

ACKNOWLEDGEMENT

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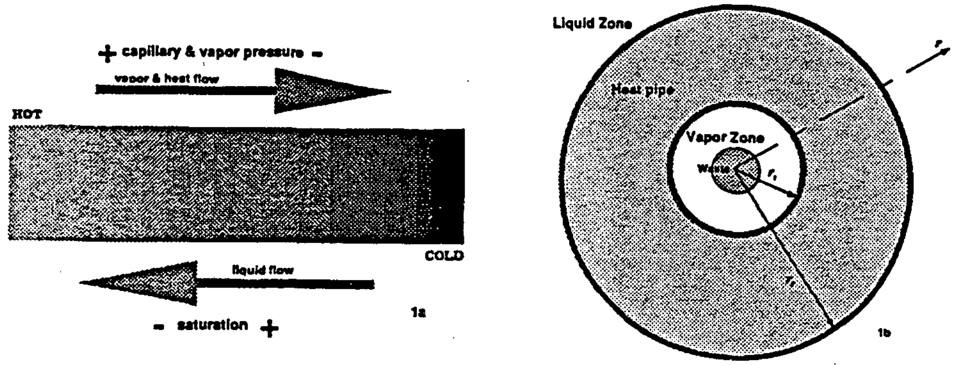

Figure 1. The Heat Pipe in Unsaturated Porous Rock.

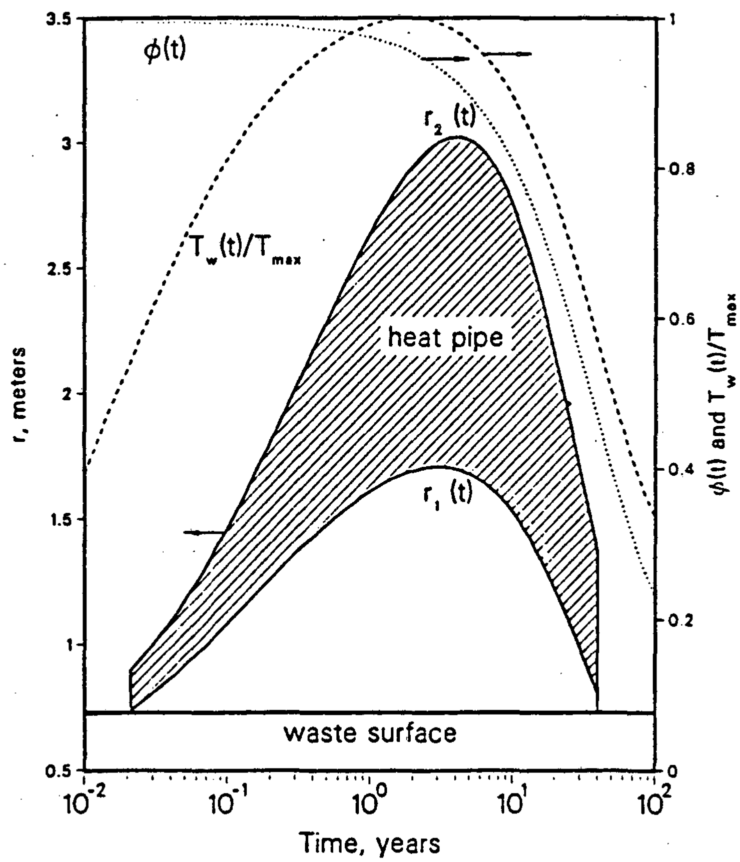

Figure 2. The Spatial Extent of a Heat Pipe as a Function of Time.

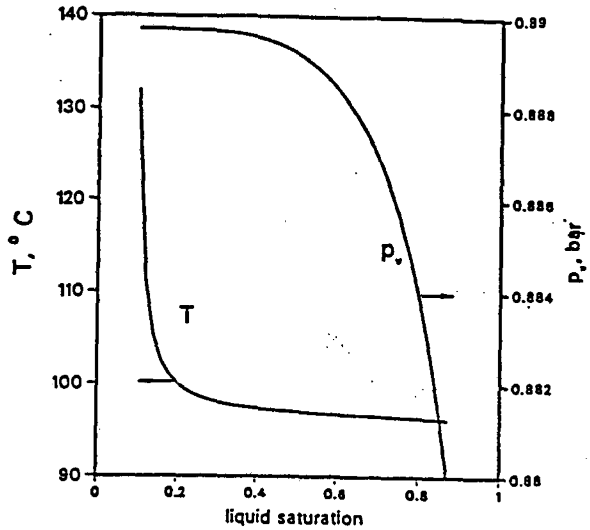

Figure 3. Temperature and Vapor Pressure Variation Within a Heat Pipe.

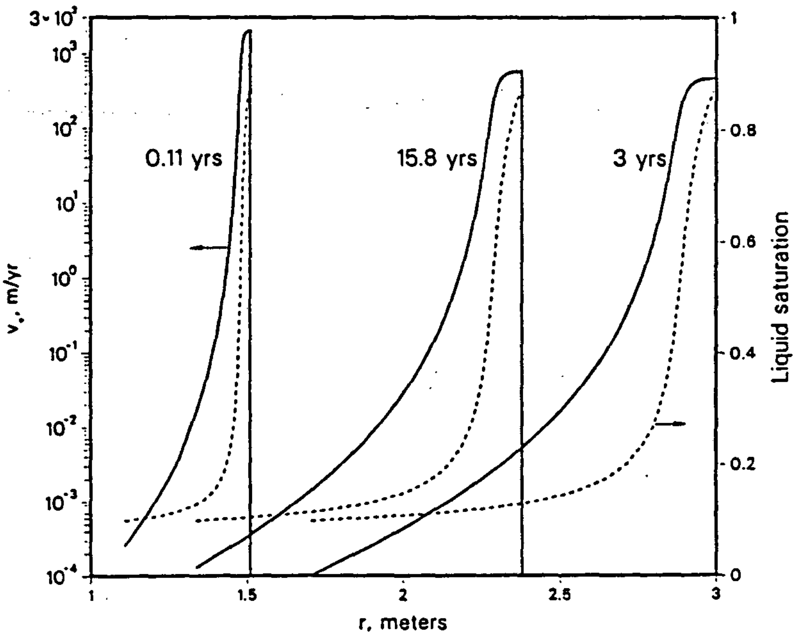

Figure 4. Vapor Velocity and Liquid Saturation in the Heat-Pipe at Various Times after Emplacement.

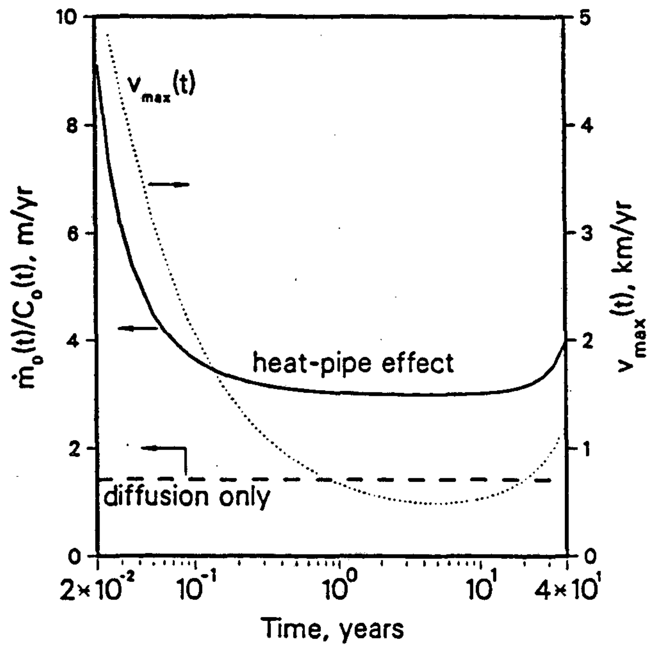

Figure 5. Normalized Mass Flux of a Gaseous Radionuclide at the Waste Surface as a Function of Time, With and Without a Heat Pipe.

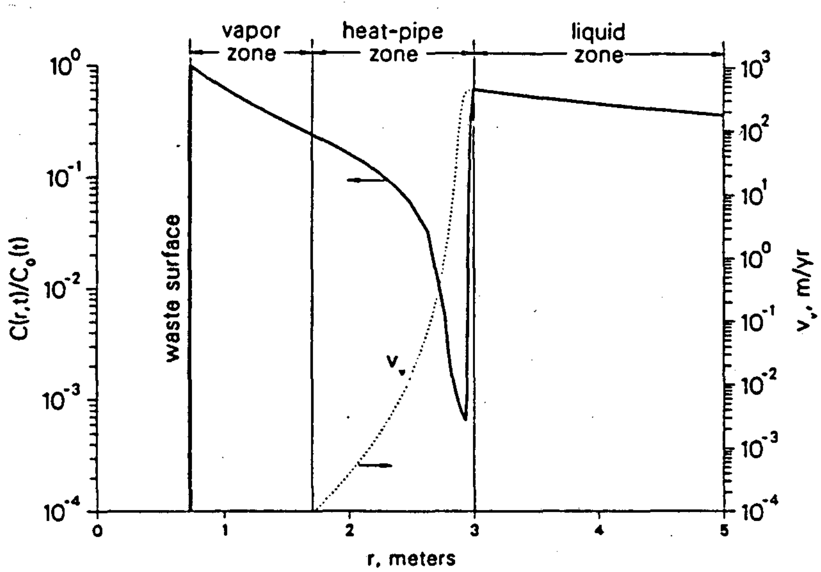

Figure 6. Concentration of a Gaseous Radionuclide, as a Function of Distance, 3 Years After Emplacement.