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February 1989



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Partially Saturated, Fractured, Porous Media**

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# ANALYTICAL MODELS FOR C-14 TRANSPORT IN A PARTIALLY SATURATED, FRACTURED, POROUS MEDIA

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## ABSTRACT

Interaction between fractures and rock matrix is considered in developing a criterion for treating fractured rock as a porous medium for the purpose of transport calculations. The value of a modified Peclet number determines the suitability of the equivalent porous medium approach. Using a porous medium model, underground concentrations of  $^{14}\text{CO}_2$  are predicted for the proposed nuclear waste repository at Yucca Mountain, Nevada. Maximum concentrations near the ground surface are comparable to the USNRC limit for unrestricted areas; travel times are predicted to be hundreds to thousands of years for the assumed parameter values.

## INTRODUCTION

The proposed nuclear repository at Yucca Mountain, Nevada would consist of sealed containers of spent fuel and other high-level waste placed in the partially saturated (or vadose) zone some 250 m above the water table and 350 m below the ground surface. Radioactive gases released from failed containers would have a relatively direct pathway to the biosphere. This presents a new problem in assessing the environmental and health impacts of such releases and in complying with regulatory standards.

We analyze the transport of  $^{14}\text{C}$  in an unsaturated, fractured, porous medium with gas-phase advection and dispersion. Transport predictions are based on a porous medium model which assumes local equilibrium between gas-phase  $^{14}\text{C}$  concentrations in the fractures and liquid-phase concentrations in the rock matrix. Justification for this assumption is based on a fracture model with advection in the fracture and transverse diffusion in the rock matrix.

Unlike Amter *et al.*<sup>1</sup> and Knapp<sup>2</sup> who have also estimated  $^{14}\text{C}$  travel-time, we (1) calculate  $^{14}\text{C}$  concentrations and fluxes, (2) include the effects of dispersion, and (3) provide a new method of justifying the local-equilibria assumptions.

## YUCCA MOUNTAIN SITE

The region around the proposed repository consists of alternating layers of welded and nonwelded tuff. The welded tuff layers are highly fractured while the non-welded layers are more sparsely fractured. Porosities are near 0.1, liquid saturation is near 0.8 and hydraulic permeabilities are very low, on the order of  $10^{-15} \text{ m}^2$ , and due mostly to fractures.

Capillary forces tend to hold vadose water in the smaller pores, leaving fractures open for gas flow. Significant gas flows have been observed under ambient conditions, leading to the expectation that, with repository heating, strong convection flows will be maintained during the thermal phase. Figure 1 shows a schematic view of the repository and the expected gas flow field. One estimate for the gas-flow velocity due to repository heating is 22 m/y in the fractures (0.04 m/y Darcy velocity) based on a simple buoyancy calculation.<sup>3</sup>

## MATHEMATICAL MODELS

We assume that  $^{14}\text{C}$  escapes from the waste containers as  $^{14}\text{CO}_2(\text{g})$ . Because carbon dioxide dissolves readily into water, we expect much of the  $^{14}\text{C}$  to be retarded by dissolution into vadose water. Some  $^{14}\text{C}$  will react to form calcite and other minerals. The amount going into these solid phases is difficult to predict and probably not significant compared to the amount in gas and liquid phases. We neglect precipitation into solid phases.

The degree to which  $\text{CO}_2$  dissolves in water is well known. We adopt, for  $^{14}\text{C}$ , the equilibrium values for  $\text{CO}_2$  in pure water at infinite dilution. Corrections for the  $^{14}\text{C}$ -isotope, capillary effects, and exact air and water compositions would be small relative to other model uncertainties. The four aqueous species  $\text{CO}_2$ , carbonic acid and bicarbonate and carbonate ions account for almost all of the liquid-phase carbon. The equilibrium values for these species allow a good approximation to be made of the ratio of  $^{14}\text{C}$  concentration in gas to  $^{14}\text{C}$

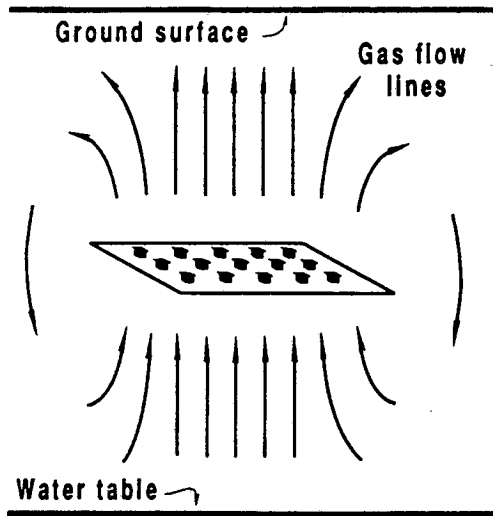


Figure 1. Schematic view of Yucca Mountain repository and gas flow field.

concentration in adjoining liquid as a function only of temperature and pH. The equilibrium values are useful because the time it takes for  $\text{CO}_2$  to equilibrate locally is short compared to other time scales of interest in this system.

We define the gas-liquid distribution coefficient for inorganic carbon to be

$$K_D = \frac{[\text{H}_2\text{CO}_3^*] + [\text{HCO}_3^-] + [\text{CO}_3^{2-}]}{[\text{CO}_2(\text{g})]} \quad (1)$$

where  $[\text{H}_2\text{CO}_3^*]$  is the sum of the aqueous  $\text{CO}_2$  and carbonic acid concentrations. The distribution coefficient can then be written in terms of the gas constant, absolute temperature, hydrogen ion concentration and three equilibrium constants as

$$K_D = RTK_H \left[ 1 + \frac{K_1}{[\text{H}^+]} + \frac{K_1K_2}{[\text{H}^+]^2} \right] \quad (2)$$

where  $K_H = [\text{H}_2\text{CO}_3^*]/p\text{CO}_2$

$$K_1 = [\text{H}^+][\text{HCO}_3^-]/[\text{H}_2\text{CO}_3^*]$$

$$K_2 = [\text{H}^+][\text{CO}_3^{2-}]/[\text{HCO}_3^-]$$

Equilibrium data<sup>5,6</sup> were used to plot the curves in Figure 2 showing  $K_D$  as a function of pH for various temperatures. Repository conditions outside the immediate container vicinities are expected to vary from 25°C to 100°C and from pH 7 to pH 9 over which  $K_D$  varies from 2 to 400. The reference value used in this paper is  $K_D = 3$  which corresponds to 50°C and pH 7.

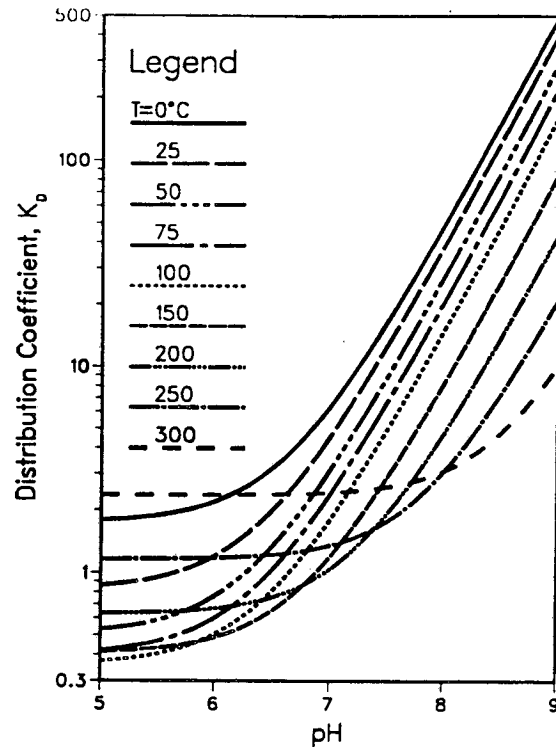


Figure 2. Equilibrium distribution coefficient  $K_D$  for carbon dioxide in water.

### Fracture Analysis

The main purpose of this model is to demonstrate for certain parameter values that local equilibrium between gas-phase carbon in the fractures and liquid-phase carbon in the rock-matrix is reasonably well attained. The tendency of the gas and liquid to equilibrate will be offset by the different flow velocities of the two phases and the distance through which some of carbon must diffuse in the liquid phase to communicate with the fractures.

We consider a single vertical planar fracture filled with gas and adjacent to a porous medium that is filled with liquid, as shown in Figure 3. Assuming that the rock matrix is fully saturated simplifies the model. Once we demonstrate local equilibrium using this model we can infer equilibrium with a partially saturated rock matrix because the diffusion coefficient in gas is several orders of magnitude higher than in liquid and equilibrium would be reached more quickly.

We neglect dispersion in the fracture and assume that the gas is well-mixed over the width of the fracture so that the concentration is uniform in the fracture in the y-direction. We also assume uniformity in the x-direction for concentrations in the fracture and rock matrix, and we assume a constant, upward gas velocity

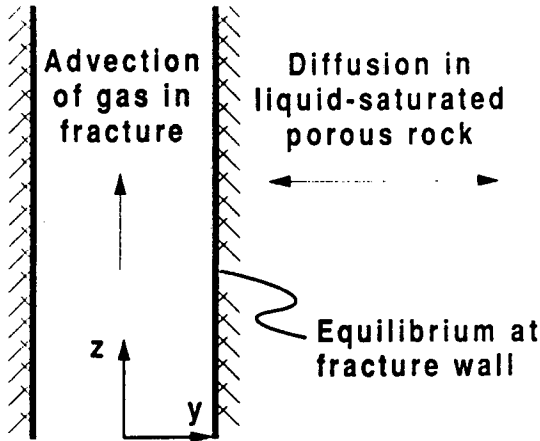


Figure 3. Fracture-model geometry showing vertical, planar fracture next to rock matrix.

in the fracture. The governing equation for the fracture domain, including the interaction term with the adjacent porous medium, is then

$$\frac{\partial C_g}{\partial t} + \lambda C_g + v_g \frac{\partial C_g}{\partial z} = \frac{\epsilon D_\ell}{b} \frac{\partial C_\ell}{\partial y} \Big|_{y=b}, \quad z > 0, t > 0 \quad (3)$$

where  $C_g$  = gas-phase concentration of  $^{14}\text{C}$   
 $C_\ell$  = liquid-phase concentration of  $^{14}\text{C}$   
 $v_g$  = velocity in the fracture  
 $\epsilon$  = total porosity in porous region  
 $D_\ell$  = effective diffusion coefficient through liquid-filled pores  
 $b$  = half width of the fracture  
 $\lambda$  = decay constant

In the porous medium we neglect advection and diffusion parallel to the fracture and keep transverse diffusion as the only transport mechanism. We treat the diffusion coefficient as a scalar constant to get the following mass balance in the rock matrix

$$\frac{\partial C_\ell}{\partial t} + \lambda C_\ell - D_\ell \frac{\partial^2 C_\ell}{\partial y^2} = 0, \quad y > b, z > 0, t > 0 \quad (4)$$

We set  $z = 0$  at the repository horizon and prescribe at that location the 'inflow' concentration in the fracture representing a gaseous release from a waste container.

$$C_g(0, t) = \psi(t), \quad t > 0 \quad (5)$$

At the fracture-wall interface where gas meets liquid we assume equilibrium between gas and liquid-phase concentrations so that

$$C_\ell(b, z, t) = K_D C_g(z, t), \quad z > 0, t > 0 \quad (6)$$

The remaining initial and boundary conditions are

$$C_g(z, 0) = 0, \quad z > 0 \quad (7)$$

$$C_\ell(y, z, 0) = 0, \quad y > b, z > 0 \quad (8)$$

$$C_g(\infty, t) = 0, \quad t > 0 \quad (9)$$

$$C_\ell(\infty, z, t) = 0, \quad z > 0, t > 0 \quad (10)$$

To make the notation more compact, we introduce the dimensionless independent variables  $Y$ ,  $Z$  and  $T$  and the decay parameter  $\Lambda$  defined

$$Y = \epsilon K_D \frac{y-b}{b}, \quad Z = \frac{z \epsilon^2 K_D^2 D_\ell}{v_g b^2} \quad (11)$$

$$T = \frac{t \epsilon^2 K_D^2 D_\ell}{b^2}, \quad \Lambda = \frac{\lambda b^2}{\epsilon^2 K_D^2 D_\ell} \quad (12)$$

At this point we consider an impulse release of  $^{14}\text{CO}_2$  flowing into the fracture at  $z = 0$

$$\psi(T) = 2C\delta(T) \quad (13)$$

with  $C$  a constant in units of concentration. We can express  $C$  in terms of the amount released by integrating the release rate over time and equating that to the total amount released  $I_o$ . The release rate is equal to the cross sectional area of the fracture,  $a$ , times the gas velocity,  $v_g$ , times the concentration,  $\psi(T)$ , leading to the equation

$$\int_0^\infty a v_g \psi dt = I_o \quad (14)$$

Using the definition of  $T$  in (12) we can change the variable of integration and substitute  $\psi(T)$  from (13) to get

$$\left( \frac{2a v_g C b^2}{\epsilon^2 K_D^2 D_\ell} \right) \int_0^\infty \delta(T) dT = I_o \quad (15)$$

or

$$C = \frac{I_o \epsilon^2 K_D^2 D_\ell}{a v_g b^2} \quad (16)$$

After solving (3)-(10), substituting the impulse release form and simplifying we get the  $^{14}\text{CO}_2$  concentration in the fracture

$$C_g(Z, T) = h(T - Z) \frac{CZ}{2\sqrt{\pi(T - Z)^3}} \exp\left[\frac{-Z^2}{4(T - Z)} - \Lambda T\right] \quad \text{for } Z > 0, T > 0 \quad (17)$$

and the liquid-phase  $^{14}\text{C}$  concentration in the matrix

$$C_l(Y, Z, T) = h(T - Z) \frac{K_D C(Z + Y)}{2\sqrt{\pi(T - Z)^3}} \exp\left[\frac{-(Z + Y)^2}{4(T - Z)} - \Lambda T\right] \quad \text{for } Y > 0, Z > 0, T > 0 \quad (18)$$

A feature of interest is the location of the peak of the concentration wave. The solution for the impulse-release case gives a smooth curve whose peak may be located by setting the partial derivative of  $C_g$  with respect to  $Z$  equal to zero. The partial derivative is zero when  $Z$  is a root of the cubic polynomial

$$Z^3 - 2(1 + T)Z^2 - 2TZ + 4T^2 = 0 \quad (19)$$

The asymptotic limit for  $t \gg b^2/\epsilon^2 K_D^2 D_l$  is given by

$$\lim_{T \rightarrow \infty} Z_{\text{peak}} \sim \sqrt{2T} \quad (20)$$

We can also define  $Y_{\text{peak}}$  as the location in the matrix where the liquid-phase concentration is maximized for given  $Z$  and  $T$  values. We obtain this also by taking a partial derivative, in this case  $C_l$  with respect to  $Y$ , and setting it equal to zero with the result for  $t \gg b^2/\epsilon^2 K_D^2 D_l$  that

$$\lim_{T \rightarrow \infty} Y_{\text{peak}} \sim \sqrt{2T} - Z \quad (21)$$

From this equation we see that the wave peak develops into a straight line in the rock matrix region with slope  $\partial Y_{\text{peak}}/\partial Z = -1$ . In dimensional terms, the magnitude of the slope  $\partial z/\partial y_{\text{peak}}$  is given by the dimensionless group which we call the modified Peclet number

$$P_l = \frac{bv_g}{K_D \epsilon D_l} \quad (22)$$

If the modified Peclet number is less than unity it means that the slope is shallow and the  $^{14}\text{C}$  wave extends well into the rock as shown schematically in Figure 4.

We also find from additional asymptotic limits of the solutions that the concentration along the wave peak is constant in the rock matrix and equal to  $K_D$  times the peak concentration in the fracture. We reason that

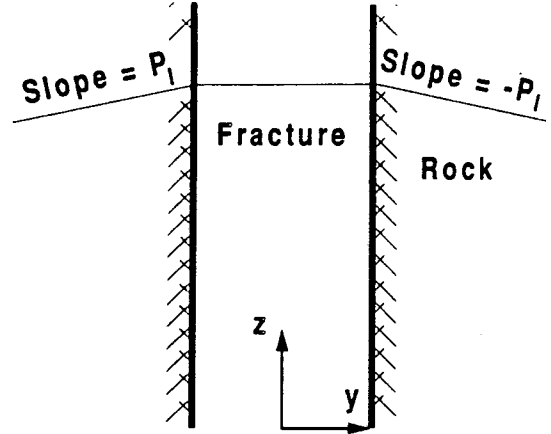


Figure 4.  $^{14}\text{C}$  penetration into the rock matrix described by the modified Peclet number. Slope of wave peak (also an isopleth) is given by  $P_l$ .

with multiple vertical fractures, the liquid concentrations in the rock matrix between fractures will be well equilibrated with gas concentrations in the fractures as long as  $P_l$  is less than unity.

#### Equivalent Porous Medium Analysis

If there is local equilibrium between  $^{14}\text{C}$  concentrations in the fractures and the intervening rock-matrix pores, we can treat the whole as an equivalent porous medium without regard to the fracture details. The following two equations describe the transport of a radionuclide in a porous medium with both gas and liquid phases

$$\left(\frac{\partial}{\partial t} + \lambda\right) \epsilon_g C_g + \nabla \cdot (\mathbf{q}_g C_g) - \nabla \cdot (\epsilon_g \mathbf{D}_g \cdot \nabla C_g) + S_{g\ell} = f_g \quad (23)$$

$$\left(\frac{\partial}{\partial t} + \lambda\right) \epsilon_l C_l + \nabla \cdot (\mathbf{q}_l C_l) - \nabla \cdot (\epsilon_l \mathbf{D}_l \cdot \nabla C_l) - S_{g\ell} = f_l \quad (24)$$

where

- $\epsilon_g$  = gas-filled porosity
- $\epsilon_l$  = liquid-filled porosity
- $\mathbf{q}_g$  = gas Darcy velocity
- $\mathbf{q}_l$  = liquid Darcy velocity
- $\mathbf{D}_g$  = dispersion coefficient in gas phase
- $\mathbf{D}_l$  = dispersion coefficient in liquid phase
- $S_{g\ell}$  = net reaction rate from gas phase to liquid
- $f_g$  = gas-phase volumetric source term
- $f_l$  = liquid-phase volumetric source term

Equations (23) and (24) can be added together to eliminate the gas-liquid reaction rate term  $S_{g\ell}$ . We then



make use of the assumption of local equilibrium between gas and liquid-phase concentrations of  $^{14}\text{C}$  by replacing  $C_\ell$  with  $K_D C_g$ . We argue in the previous section that this is a good approximation when  $P_\ell$  is less than unity. Generally the coefficients are functions of space and time. We assume here that they are constant and that the dispersion coefficients are scalar. We divide through by the group  $(\epsilon_g + \epsilon_\ell K_D)$  to get the equation

$$\left(\frac{\partial}{\partial t} + \lambda\right) C_g + \mathbf{v} \cdot \nabla C_g - D \nabla^2 C_g = \frac{f}{\epsilon_g + \epsilon_\ell K_D} \quad (25)$$

where

$$\mathbf{v} = \frac{\mathbf{q}_g + \mathbf{q}_\ell K_D}{\epsilon_g + \epsilon_\ell K_D}, \quad D = \frac{\epsilon_g D_g + \epsilon_\ell D_\ell K_D}{\epsilon_g + \epsilon_\ell K_D} \quad (26)$$

and  $f = f_g + f_\ell$  is the combined source strength.

Solutions to this equation are well known for various source terms, boundary conditions and initial conditions. We assume an infinite domain with the concentration equal to zero initially and vanishing at infinity for all time.

We consider solutions to the problems of both a point source and an infinite plane source and for an impulse, band and decaying-band release rate. Given a release-rate  $\dot{M}(t)$ , the point-source term can be written

$$f(\mathbf{r}, t) = \delta(\mathbf{r}) \dot{M}(t) \quad (27)$$

and an infinite-plane-source term can be written

$$f(\mathbf{r}, t) = \frac{1}{A} \delta(z) \dot{M}(t) \quad (28)$$

where  $A$  is the repository area normal to  $z$  over which the release rate  $\dot{M}(t)$  occurs. The decaying-band release rate is given by

$$\dot{M}(t) = \frac{I_e}{t_r} \exp(-\lambda t) [h(t - t_f) - h(t - t_f - t_r)] \quad (29)$$

where  $I_e$  is the inventory at the emplacement time  $t = 0$ ,  $h$  is the unit step function,  $t_f$  is the time between emplacement and failure and  $t_r$  is the time between failure and the end of release when the source has been exhausted. The corresponding flat-band release rate is given by the same expression without the decaying-exponent factor. The impulse release rate is obtained by taking the limit as  $t_r$  goes to zero and is written

$$\dot{M}(t) = I_e \exp(-\lambda t_f) \delta(t - t_f) \quad (30)$$

The solution for the point-source case is

$$C_g(\mathbf{r}, t) =$$

$$\int_0^t \frac{\dot{M}(\tau) \exp\left[-\frac{(\mathbf{r} - \mathbf{v}(t - \tau))^2}{4D(t - \tau)} - \lambda(t - \tau)\right]}{(\epsilon_g + \epsilon_\ell K_D) [4\pi D(t - \tau)]^{3/2}} d\tau \quad (31)$$

Similarly, the solution for the infinite-plane-source case is

$$C_g(z, t) =$$

$$\int_0^t \frac{\dot{M}(\tau) \exp[-\lambda(t - \tau)]}{(\epsilon_g + \epsilon_\ell K_D) A} G(z - v(t - \tau), t - \tau) d\tau \quad (32)$$

where

$$G(z, t) = \frac{\exp[-z^2/4Dt]}{\sqrt{4\pi Dt}} \quad (33)$$

## RESULTS

Results are based on the reference values given in Table I. Dispersion coefficient values were obtained by dividing diffusion coefficient values<sup>6</sup> by ten to account for tortuosity. The value for the fracture half-width is characteristic of reported values.<sup>7</sup> For this set of values the modified Peclet number is about 0.2. This indicates that  $^{14}\text{C}$  flowing in fractures will spread quickly into the rock matrix between fractures and thus be retarded in accordance with the local-equilibria assumptions used in the equivalent-porous-medium model.

Retardation by the liquid phase is incorporated in the advective transport velocity  $\mathbf{v}$  given by (26). This gives the speed that a  $^{14}\text{C}$  plume would travel independent of dispersion. For the reference values,  $\mathbf{v}$  is about 0.015 m/y suggesting a travel time of 2300 y for  $^{14}\text{C}$  to move from the repository to ground surface. Dispersion, however, will cause the plume to spread out and the leading edge to arrive at the ground surface more quickly.

The concentration and flux at the surface depend on the strength of the source term. The total amount of  $^{14}\text{C}$  is estimated to be 1.5 Ci/Mg U fuel at the time of emplacement.<sup>8</sup> One percent is assumed to be available for quick release independent of matrix dissolution.<sup>8</sup> With 1.5 Mg U fuel per container we arrive at 0.02 Ci for the available  $^{14}\text{C}$  inventory per failed container. For a total of 70,000 Mg U fuel in a filled repository this corresponds to about 1000 Ci of  $^{14}\text{C}$  available for quick release assuming all the containers fail. If we assume twenty percent of the containers fail,<sup>8</sup> this leaves an inventory of 200 Ci (about 40 g or 3 moles) available for quick release over the entire repository. We neglect source decay between emplacement and failure times; during the thermal period it will be unimportant.

Figure 5 shows the gas-phase concentration of  $^{14}\text{C}$  as a function of distance above a single failed container as predicted by the point-source impulse solution with a release of 0.02 Ci. Notice how the plume moves and

point-source release	0.02 Ci
plane-source release	200 Ci
plane-source area	$7 \times 10^6 \text{ m}^2$
source depth	350 m
porosity	0.1
saturation	0.8
dist. coeff. (pH 7, 50°C)	3
gas Darcy velocity	0.04 m/y
liquid Darcy velocity	0 m/y
dispersion coeff. in gas	$50 \text{ m}^2/\text{y}$
dispersion coeff. in liquid	$3 \times 10^{-3} \text{ m}^2/\text{y}$
decay constant	$1.22 \times 10^{-4} \text{ y}^{-1}$
(half life	5730 y)
gas velocity in fracture	22 m/y
fracture half width	$10^{-5} \text{ m}$

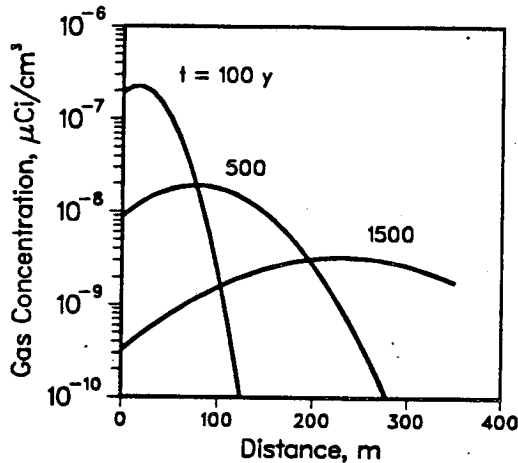


Figure 5.  $^{14}\text{CO}_2$  concentration vs. distance for impulse release of 0.02 Ci from a point source. Gas Darcy velocity is 0.04 m/y, release at time zero.

spreads out in time. By 1500 years one can see that much of the  $^{14}\text{C}$  has reached the surface (350 m) demonstrating the effect of dispersion on travel time.

The concentration in the plume decreases significantly as the plume spreads. This is attributable to dispersion because very little decay occurs in 1500 years. We conclude for this data set that dispersion has an important effect on both the travel time and the concentration at the ground surface.

Figure 6 shows the gas-phase concentration 350 meters above an infinite-plane source due to a band release of 200 Ci per  $7 \times 10^6 \text{ m}^2$  over 1000 years. The abscissa represents elapsed time from the beginning of the release. Three curves are plotted, one for the reference gas Darcy velocity 0.04 m/y, and the other two for values above and below the reference.

For the reference case, it takes about 1000 years

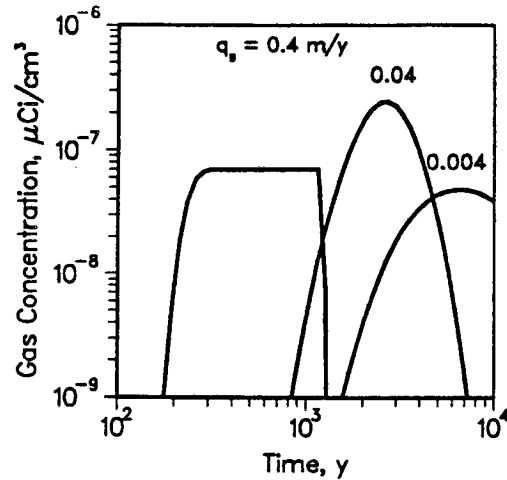


Figure 6.  $^{14}\text{CO}_2$  concentration 350 m above an infinite plane source releasing 200 Ci per  $7 \times 10^6 \text{ m}^2$  over 1000 years, release starting at time zero.

for the leading edge to reach the ground surface. The concentration peaks at a little over  $10^{-7} \mu\text{Ci}/\text{cm}^3$ , and the wave passes after about 7000 years.

With the slower velocity of 0.004 m/y, we see that it takes only slightly longer for the leading edge to reach the ground surface but that the peak concentration is lower due to decay and dispersion and that it takes much longer for the wave to pass through the ground.

When the velocity is 0.4 m/y, the curve appears cut off at the top. This is due to dilution at the source. As expected, the wave takes less time to travel to the ground surface, about 200 years, and passes through approximately 200 years after the release stops. At this flow rate, dispersion becomes less important.

Figure 7 shows what happens to the peak concentration at the ground surface when the band release duration is varied from 1 to 1000 years while the total amount released is held constant at 200 Ci. For the reference case,  $q_g = 0.04 \text{ m/y}$ , and the case  $q_g = 0.004 \text{ m/y}$  the peak concentration is not affected by the band release duration. Only at the higher gas Darcy velocity of 0.4 m/y do we see the peak concentration decreasing for long band release durations. If the release duration is shorter than the travel time, as is true where the curves are flat, dispersion will attenuate the peak of the concentration wave as it travels from the repository to the surface and tend to minimize differences caused by different release durations. If the release duration is longer than the travel time, as is true where the 0.4 m/y-curve slopes down, dispersion does not attenuate the concentration and we see the effect of the source term weakening as the release duration gets longer.

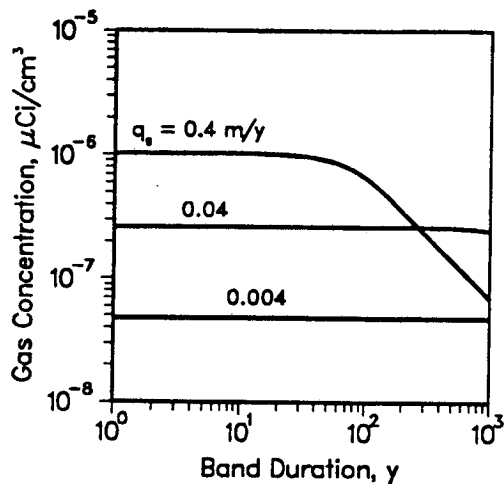


Figure 7. Peak  $^{14}\text{CO}_2$  concentration 350 m above an infinite plane source as a function of the band release duration for 200 Ci released per  $7 \times 10^6 \text{ m}^2$ .

## CONCLUSION

In the fracture analysis, we have presented a new method involving the modified Peclet number which justifies the useful assumption of local equilibria between fractures and intervening rock matrix. In the equivalent porous medium model, we have demonstrated a simple technique for estimating  $^{14}\text{C}$  transport at Yucca Mountain which requires only a small set of well-defined input parameters. We have shown that for some combinations of the input data, the concentrations and fluxes at the ground surface are independent of the source term strength. They depend on the total amount released but are not very sensitive to the rate of release.

Concentrations that we have predicted in gas approaching the ground surface are comparable to the US-NRC limit for unrestricted areas of  $10^{-7} \mu\text{Ci}/\text{cm}^3$  - Air. We have not taken into account the dilution at the ground surface as the  $^{14}\text{CO}_2$  enters the atmosphere. This will quickly lower the concentration by many orders of magnitude.

We estimate the travel time to be hundreds to thousands of years but point out that it is critically sensitive to the Darcy gas velocity and the distribution coefficient,  $K_D$ . When values for these parameters under repository operating conditions can be determined with more confidence, we will be able to better predict the behavior of  $^{14}\text{C}$  at Yucca Mountain.

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