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THE FAR FIELD MIGRATION OF RADIONUCLIDES IN TWO DIMENSIONAL GROUNDWATER FLOWS THROUGH GEOLOGIC MEDIA

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ABSTRACT

An analytical method to model the radionuclides migration in a two dimensional groundwater flow through geologic media has been developed and implemented into the computer code UCBNE21. Using this method, the potential hazard to the biosphere posed by the accidental release of radionuclides from a candidate repository site (WIPP) is determined. I-129 and Ra-226 are potentially the most hazardous nuclides in these sites but their discharge into the biosphere will not result in concentrations larger than their maximum permissible concentrations.

MIGRAÇÃO DE RADIONUCLÍDEOS EM ESCOAMENTOS BIDIMENSIONAIS ATRAVÉS DE MEIOS GEOLÓGICOS

RESUMO

Um método analítico foi desenvolvido para modelar a migração de radionuclídeos em um escoamento de águas subterrâneas bidimensionais através de meios geológicos. Este método é implementado no código de computação UCBNE21.

Utilizando este método foi determinado o risco potencial à biosfera devido à liberação acidental de radionuclídeos de um possível local nos U.S.A. para localização de repositório de rejeito de alta radioatividade (Waste Isolation Pilot Plant), em New México.

I-129 e Ra-226 são potencialmente os nuclídeos mais perigosos nestes locais mas as suas descargas para a biosfera não resultarão em concentrações maiores do que os níveis permitidos.

I - INTRODUCTION

This research is concerned with the prediction of radionuclide concentrations in aquifers in which a steady state two-dimensional groundwater flow is established. Repositories will be located in rock formations with no significant water content. However, abnormal events can occur which can cause radionuclide release from the waste package. These nuclides can eventually reach a nearby aquifer discharging into a fresh-water source.

Both migration of long actinide chains and long lived fission products in two-dimensional geological media as well as their discharges into the biosphere is modelled in this work. The theory on which this modelling is based upon was developed and presented in a previous report⁽¹⁾.

This method provides a more realistic way than a one-dimensional model to assess the potential hazard to the biosphere posed by the accidental release of radionuclides from a high-level waste repository.

Besides the capability of analyzing two dimensional flow fields, the analytical non-recursive character of the present solution to the transport equation allows one to evaluate the concentration of an arbitrary member of a chain of any length without having to solve for its precursors. The evaluation of this solution requires little computing time.

The most important result derived in the theory is that when dispersion is negligible, the solution to the one dimensional radionuclide transport equation is applicable along a streamline of the two dimensional potential flow (Darcy's flow). The problem is then divided into two parts, namely: the determination of the hydrological flow net (i.e. the potential lines and the streamlines of the flow) and the evaluation of the radionuclide concentrations along the streamlines. The discharge rate into the biosphere is also obtained.

The groundwater flow field can be represented either by analytical expressions or by (scattered) field measurements of the groundwater potentials. The analytical representation is affected by superimposing an arbitrary number of point sinks and point sources. This superimposition can represent flow fields of arbitrary complexity. On the other hand, a given number of scattered potentiometric field data can be used to represent the flow net by fitting the data with a minimal total curvature spline function⁽²⁾. The above two methods can be combined to analyze the effects of injection and pumping wells in existing aquifers.

Once the potentiometric surface is determined and values for the hydraulic conductivity at the site obtained, the streamlines and the water travel time function can be obtained by the method of characteristics. The next step is to apply the solution of the one dimensional nuclide transport equation along a streamline. Finally, if the location of the intersection of the biosphere with the contaminated streamlines is known one can evaluate the discharge rate into the biosphere.

The above described steps are performed by the computer program (UCBNE21). Due to the semi-analytical character of this program, the computing time is much smaller than finite-difference schemes used to solve the equivalent problem. A complete listing of the program is given elsewhere⁽³⁾.

As an application of the utilization of this computer program we studied the far field migration of radionuclides for two candidate repository sites. These are the Waste Isolation Pilot Plant (WIPP) in Eddy County, New Mexico and the Basalt Waste Isolation Project (BWIP) in Hanford, Washington. Hydrological data available for both sites are reviewed and the contaminated regions for the most important nuclides as well as the nuclide discharge rates, are estimated by applying this two dimensional analytical theory for radionuclide transport. Only the results for the WIPP site are presented in this paper the reader should refer to Ref. 3 for the BWIP results.

II - THEORETICAL BACKGROUND

The theoretical developments leading to the solution of the radionuclide transport equation in two-dimensional geological media are presented elsewhere⁽¹⁾. In this section we summarize the important steps shown in that work and we also emphasize the important results needed in the present work.

The two-dimensional hydrological groundwater flow is time independent, incompressible and it is a Darcy-type flow. Dispersion effects are neglected and sorption reactions are assumed

to be in equilibrium. No solubility limits are considered, but by suitably defining the boundary condition one should be able to apply the present solution with solubility limit of the first member. Water density and soil porosity are considered constants.

Subject to above assumptions, the conservation equation for the nuclide concentration in water $N_i(x,y,t)$, atoms/cm³ for the i -th member of a chain is given by

$$K_i \frac{\partial N_i}{\partial t} + \vec{\nabla} \cdot (\vec{v} N_i) = K_{i-1} \lambda_{i-1} N_{i-1} - K_i \lambda_i N_i \quad i = 1, 2, \dots \quad (1)$$

$$\lambda_0 = 0$$

Here K_i are the dimensionless retardation coefficients, \vec{v} is the groundwater velocity (m/yr), λ_i are the decay constants (1/yr). In view of the incompressibility assumption the groundwater velocity satisfies the following conservation of mass relation

$$\vec{\nabla} \cdot \vec{v} = 0 \quad (2)$$

where the groundwater velocity is given by Darcy's law

$$\vec{v} = -k \vec{\nabla} \phi ; \quad k = k' / \epsilon \quad (3)$$

Here k' is the hydraulic conductivity of the soil (m/yr), ϵ is the dimensionless porosity and ϕ is the hydraulic head (m). Although k is actually the hydraulic conductivity divided by the porosity, we will refer to k as being the hydraulic conductivity. Substituting Eq. (3) into Eq. (1) and using Eq. (2), the transport equation is rewritten as

$$K_i \frac{\partial N_i}{\partial t} - k \left(\frac{\partial \phi}{\partial x} \frac{\partial N_i}{\partial x} + \frac{\partial \phi}{\partial y} \frac{\partial N_i}{\partial y} \right) + \lambda_i K_i N_i = \lambda_{i-1} K_{i-1} N_{i-1} \quad (4)$$

$$i = 1, 2, \dots, \quad \lambda_0 = 0$$

Let us denote by R the region of interest and by P any point inside R (see Figure 1). The radionuclides are assumed to be released along a line source S in R and a point on S will be called Q . With the above notation, the initial condition and the boundary condition can be expressed by the following relationships

$$N_i(P, 0) = 0 ; \quad P \notin Q ; \quad P \in R \quad (5)$$

$$N_i(Q, t) = N_i^0 G_i(Q, t) ; \quad Q \in S, \quad t > 0 \quad (6)$$

$$G_i(P, t) \equiv 0 ; \quad t < 0, \quad (7)$$

where $G_i(Q, t)$ is an arbitrary release function which describes the time dependent dissolution and release of the waste into the aquifer, N_i^0 is the initial concentration of the i -th nuclide. Eq. (2) and Eq. (3) describe the groundwater flow field while Eq. (4), Eq. (6) and Eq. (7) govern the radionuclide transport through the geologic media.

Two-dimensional groundwater flows are representable by potential flows and one can define the streamfunction of the flow $\psi(x,y)$ by the following:

$$\frac{\partial \psi}{\partial x} = -v \frac{\partial \phi}{\partial y} \quad (8)$$

$$\frac{\partial \psi}{\partial y} = k \frac{\partial \phi}{\partial x} \quad (9)$$

The water travel time function $\sigma(x,y)$, (yr) is defined along a fixed streamline by the following equation

$$d\sigma = \frac{-d\phi}{k \left[\left(\frac{\partial \phi}{\partial x} \right)^2 + \left(\frac{\partial \phi}{\partial y} \right)^2 \right]} \quad \text{for } \psi(x,y) = \text{constant} \quad (10)$$

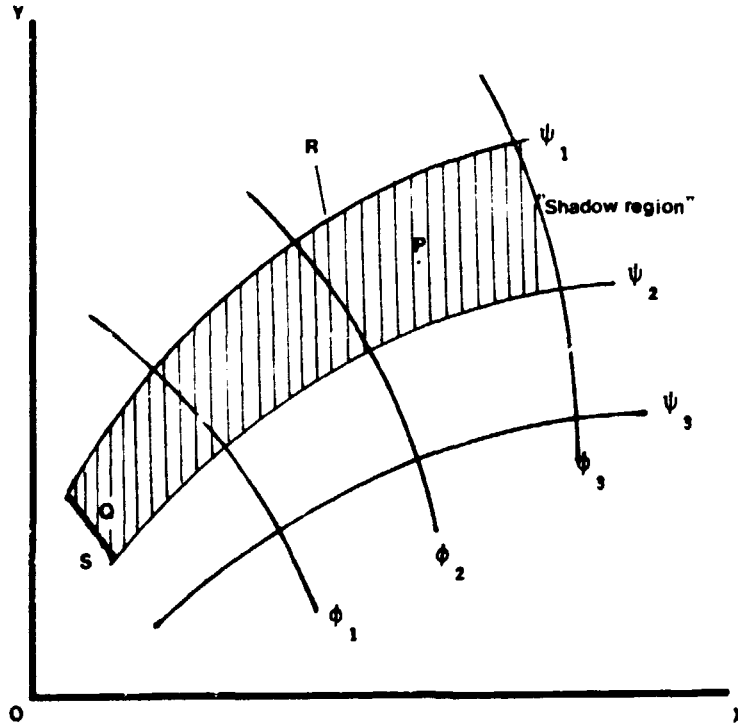


Figure 1 — Pictorial view of the system being studied: R is the region of interest, P is any point inside R, S is the source line, Q is a point on the source line, ψ_i are the streamlines and ϕ_i are potential lines.

Physically, $\sigma(x,y)$ represents the time an elementary volume of water, initially at a position (x_0, y_0) , takes to reach a given position (x,y) downstream travelling along the streamline which passes through these two points. By using the orthogonality condition between the streamfunction $\psi(x,y)$ and the potential function $\phi(x,y)$ the convective term is transformed into

$$\frac{\partial \phi}{\partial x} \frac{\partial N_i}{\partial x} + \frac{\partial \phi}{\partial y} \frac{\partial N_i}{\partial y} = \frac{\partial \phi}{\partial x} \left[\frac{\partial \phi}{\partial x} \frac{\partial N_i}{\partial \phi} + \frac{\partial \psi}{\partial x} \frac{\partial N_i}{\partial \psi} \right] + \frac{\partial \phi}{\partial y} \left[\frac{\partial \phi}{\partial y} \frac{\partial N_i}{\partial \phi} + \frac{\partial \psi}{\partial y} \frac{\partial N_i}{\partial \psi} \right] \quad (10a)$$

Finally, using the definition of the water travel time function $\sigma(x,y)$, the conservation equation for the i -th member of a radioactive chain Eq. (4) can be reduced to the following greatly simplified form

$$K_i \frac{\partial N_i}{\partial t} + \frac{\partial N_i}{\partial \sigma} + \lambda_i K_i N_i = \lambda_{i-1} K_{i-1} N_{i-1}; \quad i = 1, 2, \dots, \quad \lambda_0 = 0 \quad (11)$$

Eq. (11) is valid along a streamline where $\psi(x,y)=c$. The concentration along this streamline is now a function of only two variables, i.e. $N_i = N_i(\sigma(x,y), t)$. We will now change the notation (x,y) showing explicitly the dependence on (x,y) along a streamline by simply writing σ . Therefore, the concentration of the i -th member will be denoted by $N_i(\sigma, t)$.

Thus the two-dimensional problem of evaluating $N_i(x,y,t)$ can be reduced to a one-dimensional problem of evaluating $N_i(\sigma, t)$ along a streamline provided one knows how the water travel time varies along that streamline.

From the definition of the water travel time function Eq. (10), one can realize that lines of constant potential are parallel to lines of constant water travel time along a streamline (i.e., when $d\sigma = 0$ implies $d\phi = 0$ in Eq. (10) when $d\psi = 0$). This parallelism is mathematically expressed as

$$(\mathbf{k} \nabla \phi) \cdot (\nabla \sigma) = -1; \quad \psi(x, y) = c \quad (12)$$

The side conditions that $\sigma(x,y)$ must satisfy are (see Figure 1).

$$\sigma(Q) = 0; \quad Q \in S \quad (13)$$

$$\sigma(P) > 0; \quad P \text{ is a point in the shadow region (see Figure 1)} \quad (14)$$

The initial and boundary conditions for the radionuclide concentrations along a streamline can be restated as

$$N_i(P, 0) = N_i(\sigma, 0) = 0; \quad \sigma > 0 \quad (15)$$

$$N_i(Q, t) = N_i(0, t) = N_i^0 G_i(0, t); \quad t < 0, \quad \sigma = 0 \quad (16)$$

Let us define $G_i(t) = G_i(0,t)$. Figure 1 illustrates the system being studied. Only those streamlines which cross the source line S will be contaminated. In the case of Figure 1 all the streamlines contained between the lines ψ_1 and ψ_2 form the region which can contain radionuclides since there is no dispersion. This region downstream from the repository will be called the "shadow region". Therefore, the shadow region is the specific region of interest where the calculations are going to be performed.

For a given streamlines crossing the line source S at a point Q one must solve Eq. (11) subject to the initial condition Eq. (15) and boundary condition Eq. (16). The radionuclide concentrations are now expressed as a function of the time t started after the beginning of the release and of the water travel time σ counted from the time when the water leaves the source line S.

Mathematically, this problem can be made equivalent to its one-dimensional counterpart with a constant water velocity. Replacing (z/v) in the governing equation for the equivalent one-dimensional problem by σ , one obtains the governing equation for the two-dimensional problem valid along a streamline of the flow i.e. Eq. (11). Therefore, the solution for the one-dimensional case $N_i(z/v,t)$ can be used to express the radionuclide concentration along a streamline $N_i(\sigma,t)$ once we replace z/v by σ . The solution to the one-dimensional radionuclide transport equation with constant water velocity was developed elsewhere⁽⁴⁾. Here the solution is rewritten:

$$N_i(\sigma, t) = N_i^0 e^{-\lambda_i K_i \sigma} G_i(t - K_i \sigma) + \sum_{j=1}^{i-1} A_i^{(j)} N_j^0 \sum_{m=j}^i \frac{e^{-\lambda_m K_m \sigma}}{B_m^{(j)}} \sum_{\substack{r=j \\ r \neq m}}^i D_{rm}^{(j)} [g_{rm}(t) \otimes G_j(t)] \quad (17)$$

The coefficients are defined as follows

$$A_i^{(j)} = \prod_{r=j}^{i-1} \lambda_r K_r \quad (18)$$

$$B_m^{(j)} = \prod_{\substack{r=j \\ r \neq m}}^i (K_r - K_m) \quad (19)$$

$$D_{rm}^{(j)} = \left[\prod_{\substack{q=j \\ q \neq m \neq r}}^i (\Delta_{qm} - \Delta_{rm}) \right]^{-1} \quad (20)$$

$$\Delta_{ij} = \frac{\lambda_i K_i - \lambda_j K_j}{K_i - K_j} \quad (21)$$

The function $g_{rm}(t)$ is defined by the following expression

$$g_{rm}(t) = e^{-\Delta_{rm}(t - K_m \sigma)} h(t - K_m \sigma) \quad (22)$$

The symbol \otimes represents the convolution integral of the function in the bracketed term

$$g_{r,m}(t) \otimes G_j(t) = \int_0^t g_{r,m}(\zeta) G_j(t-\zeta) d\zeta \quad (23)$$

Repositories will be located in rock formations with no significant quantities of water, and release of radionuclides from the waste package and consequent contamination of surrounding aquifers will occur in case of an abnormal event. The proposed scenario ^(1,5) for the accidental release of radionuclides from the repository assumes that a fracture is formed which connects the repository to a surrounding aquifer. The water flowing through this fracture leaches out radionuclides from the waste package and carries them to the aquifer. Although the transport of radionuclides through this fracture takes actually some time, in this study we assume this time to be negligible. In other words, the repository is assumed to be placed directly in the aquifer being considered. This is a conservative assumption. Therefore, the source line S described in Figure 1 will indeed represent the repository.

A commonly used release mode of radionuclides from the repository is the so called decaying band release. This model states that every nuclide is released with the same constant release rate during a period called leach time T (yr). Mathematically this release mode is expressed by the following⁽⁴⁾

$$G_i(t) = \sum_{j=1}^i B_{ij} \exp(-\lambda_i t) [h(t) - h(t-T)] \quad (24)$$

where B_{ij} are the Bateman coefficients and $h(t)$ is the unit step function. The initial concentration N_i^0 is given by the following expression

$$N_i^0 = \frac{M_i^0}{QT} \quad (25)$$

here M_i^0 is the total activity (Ci) of the i-th nuclide when the leaching started, Q is the total groundwater flowrate in the aquifer through the waste (m^3/yr) and T is the leach time (yr). In this present work we adopted the above model and the solution Eq. (17) is rewritten after substituting Eq. (24) and evaluating the convolution integral

$$\begin{aligned} N_i(\sigma, t) = & N_i^0 \exp(-\lambda_i K_i \sigma) \left[\sum_{j=1}^i B_{ij} \exp(-\lambda_j(t - K_i \sigma)) \right] \left[h(t - K_i \sigma) - h(t - T - K_i \sigma) \right] \\ & + \sum_{l=1}^{i-1} \sum_{m=j}^i \sum_{r=j}^i \sum_{k=1}^l Q_{rm}^{(j)} B_{jk} \left\{ \exp \left[-\Delta_{r,m}(t - K_m \sigma) - \lambda_m K_m \sigma \right] \left[h(t - K_m \sigma) - \right. \right. \\ & \left. \left. - \exp(-T(\lambda_k - \Delta_{r,m})) h(-K_m \sigma) \right] - \exp \left[-\lambda_k t - (\lambda_m - \lambda_r \sigma) \right] \left[h(t - K_m \sigma) - h(t - T - K_m \sigma) \right] \right\} \end{aligned} \quad (26)$$

where the new coefficient is defined by

$$Q_{rm}^{(j)} = \frac{(K_r - K_m)^{i-j-1} \prod_{q=j}^{i-1} \lambda_q K_q}{(\lambda_k K_r - \lambda_k K_m - \lambda_r K_r + \lambda_m K_m) \prod_{\substack{q=j \\ q \neq r \neq m}}^i [\lambda_q K_q (K_r - K_m) + \lambda_m K_m (K_q - K_r) + \lambda_r K_r (K_m - K_q)]} \quad (27)$$

An important feature of this later form of the solution is that it allows the possibility of two or more nuclides in the same chain to have the same retardation coefficient value. The singularity caused by equal K_i values that the previous solution contained is thus removed.

If the boundary condition along every point on the source line S has the same value, this will cause the concentration along a line of constant water travel time (isochronous lines) to be also constant. Hence, the isochronous lines and the isoconcentration lines coincide with each other although each function has different values on these lines.

Therefore, to completely describe the concentration history one must obtain the groundwater flow net (i. e. the streamlines of the flow) and how $\sigma(x,y)$ varies along each streamline. When $\sigma(x,y)$ is substituted into Eq. (26), it will yield the concentration of the i-th member in the groundwater.

III - TWO-DIMENSIONAL GROUNDWATER FLOWS REPRESENTED BY AN ANALYTICAL EXPRESSION

Many groundwater flows can be adequately simulated by the superposition of an arbitrary number of point sinks and point sources in an uniform flow. In addition, when such an analytical potential function is superimposed onto the potentiometric surface of an actual aquifer (see next Section IV) one can analyse the effects of injection and/or pumping wells.

When Darcy's law Eq. (3) with constant hydraulic conductivity k is combined with the conservation of mass Eq. (2) there results

$$\nabla^2 \phi = 0 \quad (28)$$

The linear character of the above governing equation for the groundwater potential allows one to superimpose different potential fields to obtain a composite configuration. A single point source or point sink located at a position (x_j, y_j) produces a potential field in an aquifer of thickness D_0 (m), given by⁽⁶⁾

$$\phi(x, y) = \frac{-Q_j}{k \epsilon D_0 2\pi} \ln [(x - x_j)^2 + (y - y_j)^2]^{1/2} \quad (29)$$

where Q_j is the volumetric flow rate (m^3/yr) and when $Q_j > 0$, Eq. (29) represents the hydraulic head of a point source, while when $Q_j < 0$, it will represent the hydraulic head of a point sink. The uniform flow with speed v_∞ in the x-direction yields a hydraulic head given by

$$\phi(x, y) = -v_\infty x/k \quad (30)$$

Superimposing the hydraulic heads due to N distinct point sources or point sinks with an uniform flow will generate the following hydraulic head and stream functions

$$\phi(x, y) = -\frac{v_w x}{k} - \sum_{j=1}^N \frac{Q_j}{k \epsilon D_0 2\pi} \ln \left[(x - x_j)^2 + (y - y_j)^2 \right]^{1/2} \quad (31)$$

$$\psi(x, y) = -v_w y - \sum_{j=1}^N \frac{Q_j}{\epsilon D_0 2\pi} \left[\arctan \left(\frac{y - y_j}{x - x_j} \right) \right] \quad (32)$$

The velocities in the x and y directions will be respectively be given by

$$v_x = -k \frac{\partial \phi}{\partial x} = v_w + \sum_{j=1}^N \frac{Q_j}{\epsilon D_0 2\pi} \left[\frac{x - x_j}{(x - x_j)^2 + (y - y_j)^2} \right] \quad (33)$$

$$v_y = k \frac{\partial \phi}{\partial y} = \sum_{j=1}^N \frac{Q_j}{\epsilon D_0 2\pi} \left[\frac{y - y_j}{(x - x_j)^2 + (y - y_j)^2} \right] \quad (34)$$

To obtain the water travel time function along a streamline of the flow one integrates Eq. (10) with Eq. (31). However, such an integration is difficult to be carried out analytically. We will resort to the use of the method of characteristics in conjunction with a Runge-Kutta numerical integrations scheme to obtain the water travel time function. First recall the parallelism condition between the potential lines and the isochronous lines, Eq. (12)

$$\left(k \frac{\partial \phi}{\partial x} \right) \frac{\partial \sigma}{\partial x} + \left(k \frac{\partial \phi}{\partial y} \right) \frac{\partial \sigma}{\partial y} = -1; \quad d\psi = 0 \quad (35)$$

which is a first order partial differential equation (hyperbolic equation) for the variable σ , since the partial derivatives of the hydraulic head multiplied by the constant hydraulic conductivity k are known (Eq. (33) and (34)). The method of characteristics consists in determining a direction at each point of the (x, y) plane along which the integration of Eq. (35) transforms to an integration problem of an ordinary differential equation⁽⁷⁾. The detailed description on the method of characteristic will be omitted here, being sufficient to state that the characteristic equations for the Eq. (35) are given by the following

$$\frac{dx}{v_x} = \frac{dy}{v_y} = \frac{d\sigma}{-1} \quad (36)$$

The simultaneous integration of Eq. (36) starting from a point (x_0, y_0) on the initial source line S up to a point (x, y) downstream will yield the value of the water travel time at that point (x, y) . The partial derivatives of the potential function being given by Eq. (33) and Eq. (34), the above integration can be carried out by using the Runge-Kutta integration method.

IV – AQUIFERS POTENTIOMETRIC SURFACES REPRESENTED BY INTERPOLATION OF FIELD-MEASURED PIEZOMETRIC DATA

Hydrogeological studies of a proposed site for a high level waste repository usually provide an approximate description of the aquifer geometry with geologic maps and some distributed

piezometric head observations collected during field tests accumulated over the past. In addition, limited data on hydraulic conductivities are also supplied.

In our present study we use the measured piezometric data to fit a potential function. Once the potential function is determined one can solve Eq. (37) for the hydraulic conductivity k . This problem is known as the "inverse problem"⁽⁸⁾.

$$\frac{\partial}{\partial x} \left\{ k \frac{\partial \phi}{\partial x} \right\} + \frac{\partial}{\partial y} \left\{ 1 \cdot \frac{\partial \phi}{\partial y} \right\} = 0 \quad (37)$$

The problem of interpolating a smooth two dimensional function to a finite number of data points was treated by Briggs⁽²⁾. He optimizes a third order spline fit in two dimensions by minimizing the total curvature (also called the Gauss curvature) of the function. It is shown⁽²⁾ that this condition requires the desired function to satisfy the following biharmonic equation

$$\frac{\partial^4 \phi}{\partial x^4} + 2 \frac{\partial^4 \phi}{\partial x^2 \partial y^2} + \frac{\partial^4 \phi}{\partial y^4} = \phi_n \quad (38)$$

Here ϕ_n are given piezometric data points at the discrete points $(x_n, y_n), n = 1, 2, \dots, M$. Numerical solution to Eq. (38) when written as a set of algebraic equations by expressing the derivatives with finite differences yields the desired function $\phi(x, y)$. This method has been programmed into the Surface Gridding Library (SGL) and implemented at the LBL computer center⁽⁹⁾.

Therefore, by specifying a given number of values for the potential function at different points inside the region of interest one can construct the potentiometric surface by using the SGL computer program. Once the potential function is known, the next step is to solve the inverse problem, i.e. to determine the hydraulic conductivity k .

Different solutions have been proposed to the inverse problem. Among those, Emsellem and Marsily⁽¹⁰⁾ propose a modified form of the Cauchy's problem, Frind and Pinder⁽¹¹⁾ propose a Galerkin's scheme solution, Nelson⁽¹²⁾ makes use of the energy-dissipation method while Sagar⁽¹³⁾ solves algebraically the finite difference form of the Eq. (37). These solutions require either complex numerical schemes or assumptions which might bias the original data.

In this present work the method of characteristics is used to solve the Cauchy's problem. Consider Eq. (37) where $\phi(x, y)$ is known. Eq. (37) can be rewritten as a first order differential equation (hyperbolic equation) in the unknown variable $k(x, y)$

$$m(x, y) \frac{\partial k}{\partial x} + n(x, y) \frac{\partial k}{\partial y} = -k \quad (39)$$

where $m(x, y)$ and $n(x, y)$ are known functions given by the following

$$m(x, y) = \frac{\partial \phi / \partial x}{\nabla^2 \phi} ; \quad n(x, y) = \frac{\partial \phi / \partial y}{\nabla^2 \phi} \quad (40)$$

It can be shown⁽⁷⁾ that integration of Eq. (39) in the region of interest R is possible when boundary conditions for $k(x, y)$ are prescribed along a line S cutting every streamline of the flow in the region R i.e. when a value of k is known on every streamline of the region R .

Since the potential function $\phi(x,y)$ has already been evaluated by SGL, the streamline can be determined by using the method of characteristics. So defined the problem of finding the hydraulic conductivity can be solved by the method of characteristics and possess an unique solution⁽⁷⁾. Since the governing equation for the hydraulic conductivity has the same form (hyperbolic equation) as the governing equation for the water travel time function Eq (35) one can readily obtain the characteristic equations

$$\frac{dx}{m(x, y)} = \frac{dy}{n(x, y)} = \frac{dk}{-k} \quad (41)$$

In order to integrate Eq. (41) one needs therefore to specify values for the hydraulic conductivity k along the source line S . The integrations are carried out numerically using a Runge-Kutta scheme. The partial derivatives of the potential function are expressed in finite central difference form.

V – EVALUATION OF RADIONUCLIDE DISCHARGE RATES INTO THE BIOSPHERE AND CUMULATIVE DISCHARGE IN THE BIOSPHERE

Determination of $N_i(x,y,t)$ provides us with a quantitative picture of the migration of that particular nuclide through the geologic media after being released from the repository. However, one measure of the potential hazard to the general public can be obtained by evaluating the cumulative amount of curies of individual nuclides actually discharged into a potential source of fresh water, taking into account decay after discharge.

Let us denote by $q_i(t)$ the discharge rate (Ci/yr) of the i -th member of a radionuclide chain into the biosphere at a time t after chain was released from the source line. Consider the intersection of the "shadow region" streamlines with the biosphere which is represented by a curve B . This curve is subdivided into n subcurves of arc with length ds_k as shown in Figure 1a.

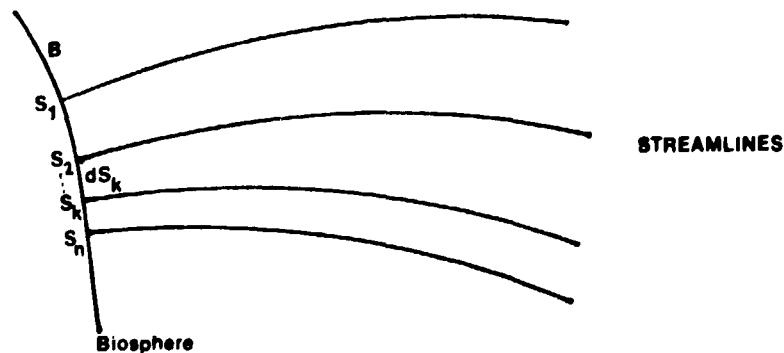


Figure 1a. – Pictorial view of the intersection of the contaminated streamlines with the biosphere.

Let the water velocity at a point s_k be $v(s_k)$, (m/yr), the aquifer thickness be $h(s_k)$, (m) and the nuclide concentration $N_i(\sigma(s_k), t)$. The discharge rate through an element ds_k is given by

$$dq_i = v(s_k)h(s_k)N_i(\sigma(s_k), t) ds_k \quad (42)$$

To obtain the discharge rate for all the contaminated streamlines that reach the biosphere one integrates Eq. (42) along the entire curve B

$$q_i(t) = \int_{s_1}^{s_n} v(s_k)h(s_k)N_i(\sigma(s_k), t) ds_k \quad (43)$$

The concentration $N_i(\sigma(s_k), t)$ is readily evaluated by using Eq. (26). The water velocity is obtained by the following expression

$$v(s_k) = [v_x^2 + v_y^2]_{s_k}^{1/2} \quad (44)$$

However, the aquifer thickness at the biosphere $h(s_k)$ is generally not known. To avoid this problem we make use of the fact that the volumetric flow rate between two streamlines is constant throughout the field. Since the product $v(s_k)h(s_k)ds_k$ is the volumetric flow rate between the streamlines passing through two points along the curve B, this is equal to the volumetric flow rate at the source line between corresponding streamlines. Aquifer thickness at the repository is generally better known than in the far field because more extensive geological studies are carried near the repository site than in the far field. This allows one to evaluate the volumetric flow rate between those streamlines.

The integral in Eq. (43) is readily evaluated by a proper numerical scheme (Romberg method⁽¹⁴⁾) once the integrand is determined for a number of points along the curve B.

The cumulative discharge of a i -th member of a radionuclide chain, $Q_i(t), (Ci)$ is obtained by integrating the discharge rate $q_i(t)$ from the first nuclide arrival time up to a time t afterwards. The integral is multiplied by the Bateman function to account for the decay of the members of the chain after they are discharged. The radionuclides are assumed to remain stationary in a reservoir after their discharge.

$$Q_i(t) = \int_{t_a}^t q_i(\tau) \sum_{j=1}^i B_{ij} \exp(-\lambda_j(t-\tau)) d\tau \quad (45)$$

The first nuclide arrival time t_a , (yr) is an important quantity in assessing the potential hazard of a particular radionuclide. It can be evaluated by the following formula

$$t_a = \sigma_{\min} K_{\min} \quad (46)$$

Here, σ_{\min} , (yr) is the smallest value of the water travel time on the biosphere curve and K_{\min} is the smallest retardation coefficient of all the precursors in the chain. If the chain has only one member, K_{\min} will be its own retardation coefficient.

In order to be able to compare the potential hazard due to the discharge of different radionuclides into the biosphere one can use the water dilution rate $w_i(t)$, (m^3/yr) as defined in Reference 1. The water dilution rate is obtained by dividing the discharge rate by the Maximum Permissible Concentration, MPC given in (Ci/m^3) by 10CFR20. Thus

$$w_i(t) = q_i(t)/MPC_i \quad (47)$$

This ratio is the necessary water flow rate in the biosphere so that it will dilute the discharge rate of that nuclide to drinking water standards. This calculation provides an additional advantage since one can add these required flow rates for every nuclide to obtain an overall hazard index for all nuclides present in the repository.

Similarly, if one divides the cumulative discharge $Q_i(t)$ by the MPC of that i-th member one obtains the total amount of water that is needed at the biosphere to dilute the cumulative amount of that nuclide to drinking water standards. This quantity is called water dilution volume⁽¹⁾ and is expressed as follows

$$W_i(t) = Q_i(t)/MPC_i \quad (48)$$

Finally, the total water dilution rate is the sum of all water dilution rates of individual nuclides and the total water dilution volume is, in the same way, the sum of water dilution volumes of individual nuclides discharging at a given time.

VI – MODELLING OF THE FAR FIELD RADIONUCLIDE MIGRATION AT THE REFERENCE SITE OF THE WASTE ISOLATION PILOT PLANT (WIPP) IN EDDY COUNTY, NEW MEXICO

Hydrological data used in this work were obtained primarily from the Department of Energy report: Waste Isolation Pilot Plant⁽¹⁵⁾ which summarizes the data reviewed by several authors. Among these are Mercer and Orr⁽¹⁷⁾, Register⁽¹⁶⁾ and others. In this report we present a brief summary of the available information and the reader should refer to above references for more detailed descriptions of the hydrology in the region of the WIPP site.

There are two potential pathways for the radionuclides to reach the biosphere after they are released from the repository. The aquifers contained in the Rustler formation overlying the Salado formation flow southwestward and discharge into Pecos River in the vicinity of Malaga Bend. On the other hand, underlying the repository site we have the Delaware Mountain Group aquifers which contrary to Rustler aquifers flow northward discharging into the Capitan aquifer which is a source of fresh water.

Two sets of hydraulic potentials representing the Rustler aquifer are given in Ref. 15. One for the measured hydraulic head and the other for the calculated hydraulic head. Calculations are done using both set of hydraulic heads and comparisons are drawn. In the same way, two sets of hydraulic heads for the underlying Delaware Mountain Group aquifers are presented in Ref. 15. Data on measured aquifer parameters in the region are not extensively available and the hydraulic conductivity data show a large range of variation. However, hydraulic conductivity at the repository site can be estimated^(16,17). A summary of the hydrological data at the site used in this work is given in Table I.

The Salado formation where the repository will be located and the underlying Castile formation are essentially impermeable and contain no water. In this present analysis we postulate that any radionuclide accidentally released from the waste package due to any abnormal event is transferred through an open fracture to either the overlying aquifers in the Rustler formation or to the underlying Delaware Mountain Group aquifers.

Table I

Summary of Hydrological Data at the (WIPP) Repository Site (15,16)

	Rustler aquifer	Delaware aquifer
Thickness (m)	65	900
Hydraulic conductivity (m/yr)	A range from 0.1 to 140; used Value = 11.	0.3
Porosity	0.10	0.16

Although the transport of radionuclides in groundwater through these fractures might take some time, we assume this time to be negligible when compared to the transport time in the aquifer itself (typical values are ten years compared to 10000 years). Therefore, conceptually, the model repository is located in the aquifer being studied. This is a conservative assumption because, if the travel time in the fracture was to be considered, the arrival time of the first nuclide would be larger and also, the concentrations would be smaller.

In assessing the safety characteristic of the site, the Rustler formation aquifers are of primary importance because the hydraulic head in the Delaware aquifer being larger than that of the Rustler aquifer, produces an upward driving gradient. In addition, thermally induced convective flows make the downward seepage of groundwater unlikely to occur. However, we calculated the migration of radionuclides in both aquifers for the sake of completeness.

The contaminated regions of the most hazardous nuclides (e.g. I-129, Ra-226) for different times after the leaching started are determined by using the computer code UCBNE21⁽⁵⁾ developed based on the model presented above.

The important result derived from these calculations are the time of contaminant arrival to the biosphere and the location of arrival. In addition, once the location and time of contaminant arrival are known, one can evaluate the discharge rate as well as the cumulative discharge into the biosphere. These quantities present a quantitative measure of the efficiency of the repository as well as of the geologic media as barriers for the radionuclide migration.

All the long lived fission products and the four long actinide chains⁽¹⁾ are considered in this calculation, although only the results for the most hazardous ones will be reported. Several nuclides can be discarded as potentially hazardous because they either have a small half life (with no long lived precursors), a large retardation coefficient, small toxicity, small initial activity or a combination of these possibilities. To rank the potential hazard of each nuclide due to its discharge into the biosphere let us consider the following group of parameters

$$10T_{1/2}/K_{\min} \quad \text{and} \quad 0.001M_i^0/(MPC)_i \quad (49)$$

Physically, the first group of parameters represents the water travel time if the nuclides were to arrive at the biosphere after 10 half lives. Therefore, if the calculated water travel time at the biosphere is equal to or larger than the first parameter in Eq. (49), the concentration of that nuclide has already decayed to a fraction 0.001.

This leads us to the meaning of the second group of parameters. M_i^0 are the initial activities of each nuclide at the time of burial and therefore $M_i^0/(MPC)_i$ represents the volume

of water required to dilute that activity to drinking water standards. Hence, the second parameter in Eq. (49) is the water volume required to dilute the activity present after ten half lives, which was arbitrarily chosen to yield 0.1% of the original activity. For shorter lived actinides (e. g. Ra-226) we consider the parameters of its controlling precursor (e. g. U-234 for times scales of the order of 10^5 years or U-238 for longer time scales) and one assumes Ra-226 to be in secular equilibrium with either U-234 or U-238 because the time scale is large enough. Therefore, the potential hazard a nuclide poses to the biosphere can be measured by the first group of parameters ($10T_{1/2}/K_{min}$) which represents the migration characteristic of that nuclide and by the second group of parameter ($0.001M_i^0/MPC_i$) which represents the toxicity and the total amount of that nuclide initially present. By considering the product of the two group of parameters one can obtain an overall hazard index which can be used to compare different radionuclides in order to rank them.

$$I_i = (10T_{1/2}/K_{min}) \ln(0.001M_i^0/(MPC)_i) \quad (50)$$

The long lived fission products parameters are listed in Table II and the actinide chains parameters in Table III. By comparing the product of the two parameters in Eq. (50) one can rank all nuclides as far as potential hazard to the public goes. This ranking is shown in Table IV. Ra-226 is ranked first because its controlling parent, U-238, requires a water travel time of about 10^9 years to decay ten half lives. The next ranked nuclide is I-129 which, due to its retardation coefficient of unity, requires a water travel time of 10^8 years to decay ten half lives. As our calculation will show, the next ranked nuclides Tc-99, U-234, etc. contribute very little to the total discharge rate (expressed in terms of total water dilution rate).

Table II

Long Lived Fission Products Parameters for WIPP Site

Nuclide	$T_{1/2}$ (yr)	K_i (1)	$(MPC)_i$ (2) (Ci/m ³)	M_i^0 (3) (Ci/GwYr)	$10T_{1/2}/K_i$ (yr)	$.001M_i^0/MPC$ (m ³ /GwYr)
H-3	1.2E+1	1.0E+0	3.0E-3	1.9E+4	1.2E+2	6.3E+3
C-4	5.6E+3	1.0E+1	8.0E-4	1.3E+3	5.6E+3	1.6E+1
Se-79	6.5E+4	6.0E+2	3.0E-4	1.1E+1	1.1E+3	3.6E+1
Sr-90	2.8E+1	1.0E+0	3.0E-7	2.1E+6	2.8E+2	7.0E+8
Zr-93	9.5E+5	1.0E+4	8.0E-4	5.2E+1	9.5E+2	6.4E+1
Nb-93m	1.4E+1	1.0E+4	4.0E-4	5.0E+0	1.4E-2	1.2E+1
Tc-99	2.1E+5	1.0E+0	2.0E-4	3.9E+2	2.1E+6	1.9E+3
Ru-106	1.0E+0	1.0E+1	1.0E-5	1.1E+7	1.0E+0	1.1E+9
Cd-113m	1.4E+1	1.0E+4	3.0E-5	1.3E+3	1.4E-3	4.5E+4
Sn-126	1.0E+5	1.0E+3	2.0E-7	1.5E+1	1.0E+3	7.4E+2
Sb-125	2.7E+1	1.0E+2	1.0E-4	2.1E+5	2.7E+0	2.1E+6
I-129	1.7E+7	1.0E+0	6.0E-8	1.0E+0	1.7E+8	1.7E+4
Cs-137	3.0E+1	4.1E+2	2.0E-7	2.9E+6	7.3E-1	1.5E+8
Cs-135	3.0E+6	4.1E+2	1.0E-4	7.8E+0	7.3E+4	7.8E+1
Ce-144	7.8E-1	1.2E+4	1.0E-7	2.1E+7	6.5E-4	2.1E+9
Pm-147	4.4E+0	2.5E+3	2.0E-4	2.6E+6	1.8E-2	1.3E+7
Sm-151	8.7E+1	2.5E+3	4.0E-4	3.4E+7	3.5E-1	8.5E+4
Eu-152	1.3E+1	2.5E+3	6.0E-7	3.3E+2	6.1E-1	5.5E+3
Eu-154	1.8E+1	2.5E+3	2.0E-5	1.9E+5	6.4E-2	9.4E+6
Eu-155	1.8E+0	2.5E+3	2.0E-4	1.7E+5	7.2E-3	8.7E+5

(1): From reference 15.

(2): From 10CFR20, Appendix B and 15.

(3): For reprocessed high level waste, LWR, 10 years decay, 0.5% U and Pu loss into the waste, 15.

Table III

Actinide Chains Parameters for the WIPP Site

Nuclide	$T_{1/2}$ (yr)	K_i (1)	(MPC) $_i$ (2) (Ci/m ³)	M_i^0 (3) (Ci/GwYr)	$10T_{1/2}/K_i$ (yr)	$.001M_i^0/MPC$ (m ³ /GwYr)
Ra-225	4.1E-3	6.8E+2	5.0E-7	—	1.1E+3	8.7E+5
Ra-226 ***	1.6E+3	6.8E+2	3.0E-8	—	8.9E+4	1.5E+3
Ra-226 ****	1.6E+3	6.8E+2	3.0E-8	—	1.6E+9	5.5E+1
Th-228	1.9E+0	5.9E+4	7.0E-8	—	1.1E+0	8.9E+4
Th-229	7.3E+3	5.9E+4	5.0E-7	—	1.1E+3	1.2E+4
Th-230 ***	8.0E+4	5.9E+4	2.0E-6	—	8.9E+4	3.2E-1
Th-230 ****	8.0E+4	5.9E+4	2.0E-6	—	1.6E+9	8.4E-3
U-233	1.6E+5	2.8E+1	3.0E-5	—	1.1E+3	2.0E+2
U-234	2.5E+5	2.8E+1	3.0E-5	1.2E+0 *	8.9E+4	3.8E+1
U-235	7.1E+8	2.8E+1	3.0E-5	2.3E-3	2.5E+8	7.7E-2
U-238	4.5E+9	2.8E+1	4.0E-5	4.3E-2	1.6E+9	1.0E+0
Np-237	2.1E+6	1.9E+4	3.0E-6	1.5E+1 **	1.1E+3	5.2E+3
Pu-238	8.6E+1	5.7E+4	5.0E-6	5.1E+2	1.5E+2	1.0E+5
Pu-239	2.4E+4	5.7E+4	5.0E-6	4.4E+1	4.2E+0	8.8E+3
Pu-240	6.6E-3	5.7E+4	5.0E-6	2.7E+2	1.1E-0	5.3E+4
Pu-241	1.3E+1	5.7E+4	2.0E-4	1.4E+4	3.1E+1	7.0E+4
Pu-242	3.8E+5	5.7E+4	5.0E-6	1.9E-1	6.6E+1	3.8E+1
Am-241	4.5E+2	1.0E+4	4.0E-6	4.9E+3	4.6E-1	1.2E+6
Am-242m	1.5E+2	1.0E+4	4.0E-6	1.2E+2	1.5E-1	2.9E+4
Am-243	7.9E+3	1.0E+4	4.0E-6	4.8E+2	7.9E+0	1.2E+5
Cm-242	4.5E-1	3.0E+3	2.0E-5	4.4E+5	1.5E-1	2.2E+7
Cm-243	3.2E+1	3.0E+3	5.0E-6	9.0E+1	1.0E-1	1.8E+4
Cm-244	1.8E+1	3.0E+3	7.0E-8	7.4E+4	5.9E-2	1.0E+7
Cm-245	9.3E+3	3.0E+3	4.0E-6	9.8E+0	3.1E+1	2.4E+3
Cm-246	5.5E+3	3.0E+3	4.0E-6	1.9E+0	1.8E+1	4.8E+2

* Includes the decay of Pu-238, Am-242m and Cm-242.

** Includes the decay of Pu-241 and Am-241.

*** Based on U-234 value

**** Based on U-238 value

(1): From reference 15.

(2): From 10CFR20, Appendix B and 15.

(3): For reprocessed high level waste, LWR, 10 years decay, 0.5% U and Pu loss into the waste, 15. Am-242m is in the metastable form.

Table IV

Potential Hazardous Nuclide Ranking for the WIPP Site

Ranking	Nuclide	$10T_{1/2}/K_i$	I_i
1	Ra-226	1.6E+9 *	6.5E+9
2	I-129	1.7E+8	1.7E+9
3	Tc-99	2.1E+6	1.6E+7
4	U-234	8.9E+4	3.2E+6
5	Cs-135	7.3E+4	3.2E+6
6	Se-179	1.1E+3	2.3E+4

*Based on the U-238 value

The calculated potentiometric data for the Rustler aquifer¹⁵ is represented in a three dimensional plot in Figure 2. The projection of the Pecos River flow pattern onto the potentiometric surface is also shown. The shape of the surface clearly indicates that the general direction of flow is southward. The presence of a "valley" in the potentiometric surface will cause the streamlines to converge in that region. Such valleys are caused either by a strong sink, as in the case of a river, or by a region of high hydraulic conductivity, as in the case of fracture zones. In the case of the Rustler aquifer with the calculated potentiometric data it seems that the valley in the potentiometric surface near Malaga Bend is caused by both the river and a large value of the hydraulic conductivity.

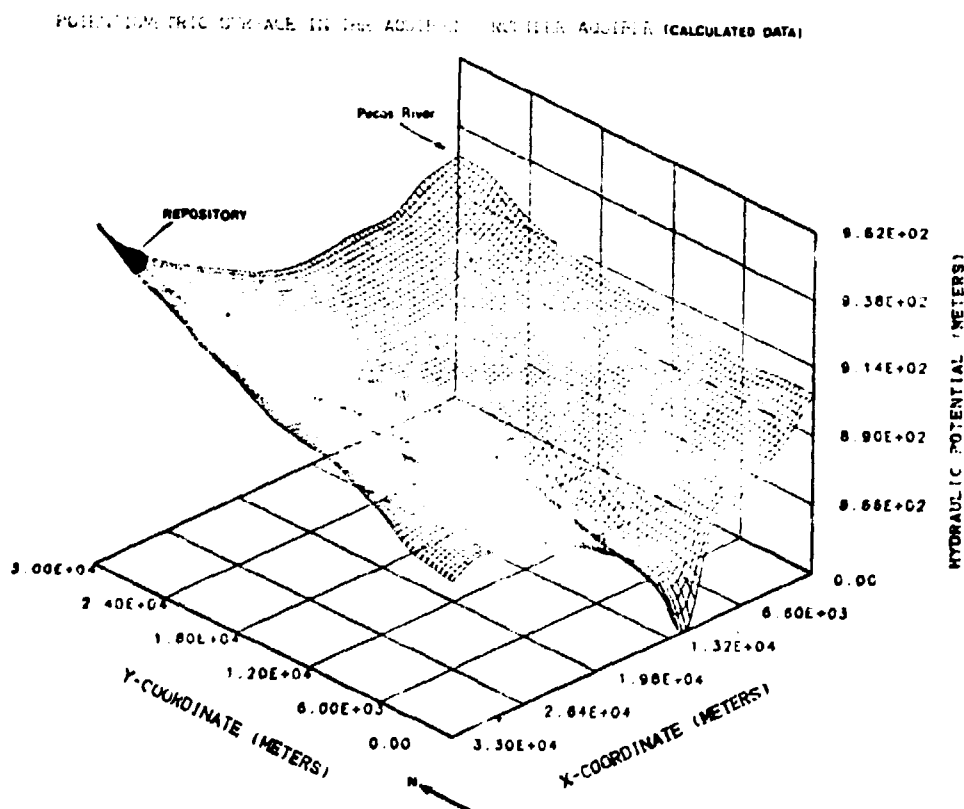


Figure 2 - The potentiometric surface (calculated data) in Rustler aquifer with the projection of Pecos River onto the surface.

Hydrological calculations for the determination of the contaminated streamlines, hydraulic conductivities and water travel times were performed for both sets of potentiometric data. The entire shadow region is covered by five streamlines in each case. The water travel time to reach the biosphere (Pecos River) for each streamline for each set of the potentiometric data is shown in Table V.

Table V

Water Arrival Time (years) at the Biosphere for the WIPP Site

Streamlin nº	Rustler Aquifer (calculated data)	Rustler Aquifer (raw data)	Delaware Aquifer (raw data)
1	2.23E+4	2.17E+4	1.68E+6
2	3.15E+4	9.73E+3	6.24E+6
3	4.65E+4	1.82E+4	1.80E+6
4	4.24E+4	3.33E+5	1.40E+6
5	4.11E+4	3.50E+5	3.46E+6

For the calculated potentiometric data set the shortest water travel time to the biosphere is 2.23E+4 years, while for the raw data set the shortest water travel time to the biosphere is 9.73E+3 years. From these values one can conclude that only the first five ranked nuclides in Table IV will reach the Pecos River after migrating through Rustler aquifer. All other nuclides will be retarded and they will have decayed before reaching the Pecos River.

Figure 3 shows the I-129 contaminated region indicated by the dotted areas at 10^4 and 10^5 years after the leaching started. The lines of constant potential are labelled with their values expressed in meters above mean sea level. Only the streamlines of the repository shadow region are shown. For this figure the calculated potential data were used. As was expected, the I-129 will discharge into Pecos River near Malaga Bend. After 10^5 years almost all I-129 have already been discharged into Peco River.

Figure 4 shows the water dilution rate of I-129 in the Pecos River for the calculated set of potentiometric data. Three values of leach time were used for comparasions (10^4 , 10^5 and 10^6 years). Increasing the value of the leach time by an order of magnitude decreases the water dilution rate by approximately one order of magnitude for I-129. The first nuclide arrival time is about 2.0E+4 years for I-129.

Similarly, Figure 5 shows the water dilution rate and water dilution volume for Ra-226 discharging into Pecos River from Rustler aquifer with the calculated set of potentiometric data. The peak water dilution rate and the peak water dilution volume are three orders of magnitude smaller than for I-129 and the first arrival time for Ra-226 is 8.0E+5 years.

The calculations were repeated for the other nuclides ranked in Table IV but their contribution to the total water dilution rate were negligible when compared to I-129 and Ra-226 contributions. Therefore, the total water dilution rate before 8.0E+5 years is due primarily to I-129 and after that time Ra-226 becomes the major contributor but with a peak value three orders of magnitude smaller. In addition to these quantitative considerations one should notice that the location of discharge for the contaminated streamlines into the Pecos River is practically localized.

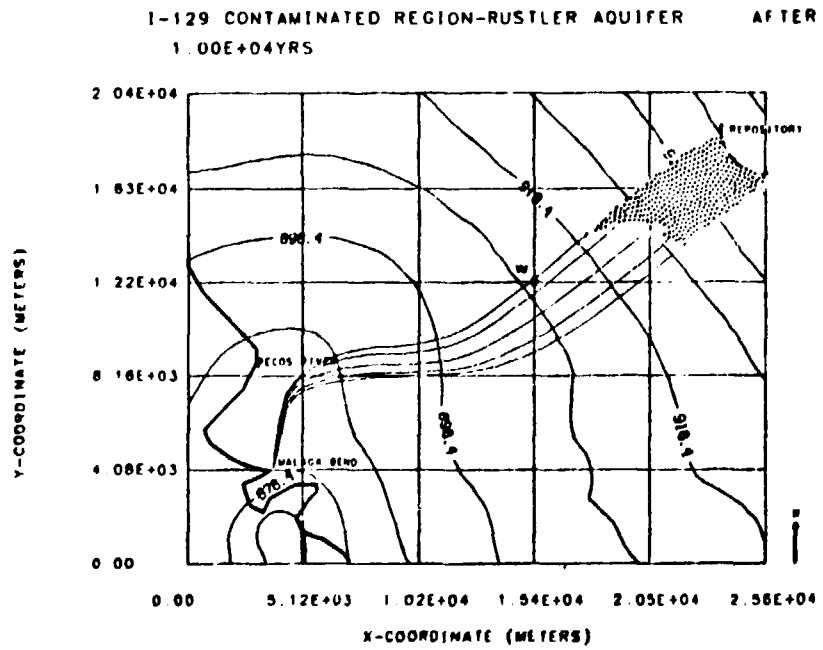
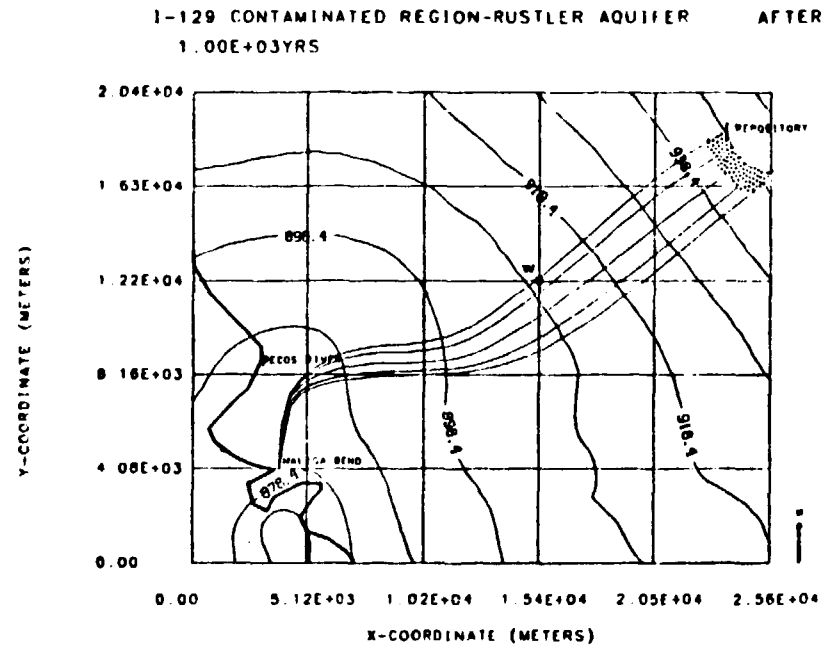


Figure 3 - 1-129 contaminated region in Rustler aquifer (calculated data after 10^3 and 10^4 years. Potentials are in meters above MSL.

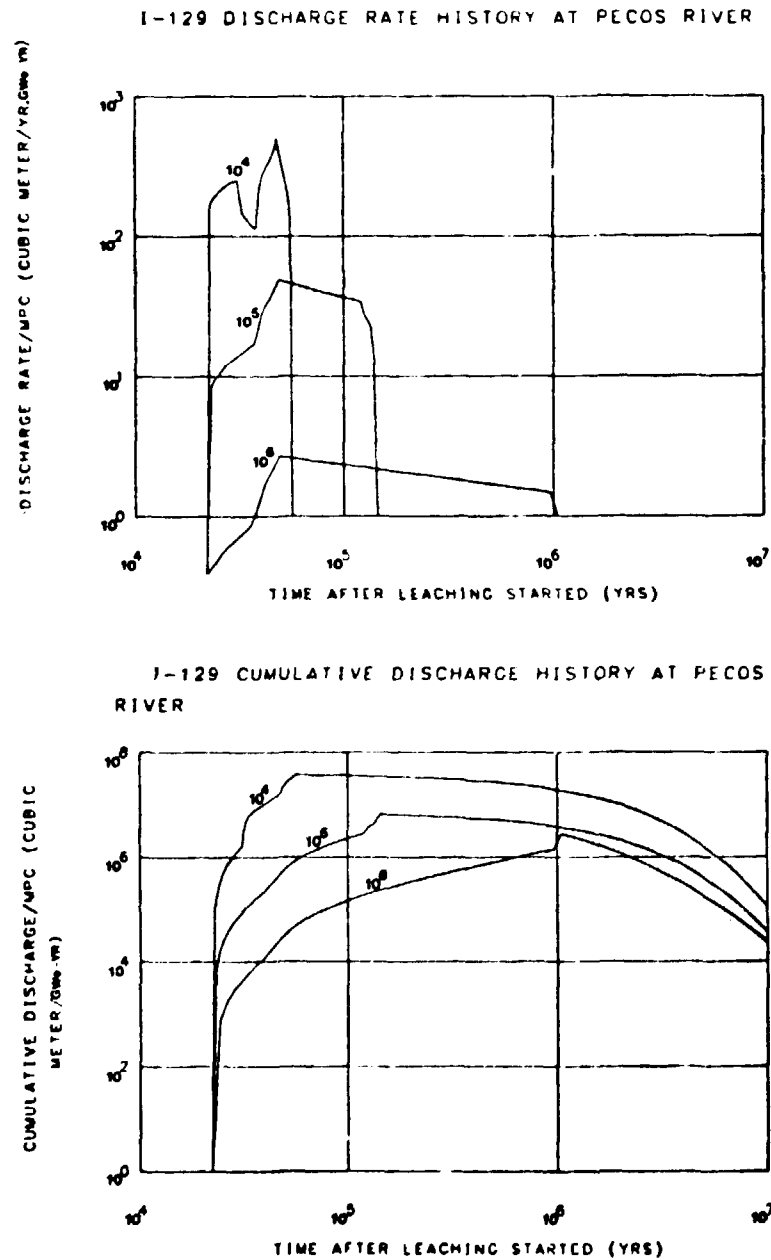


Figure 4 - I-129 discharge rate/MPC (water dilution rate) and cumulative discharge (water dilution volume) at Pecos River discharging from Rustler aquifer (calculated data) for leach of 10^4 , 10^5 and 10^6 years.

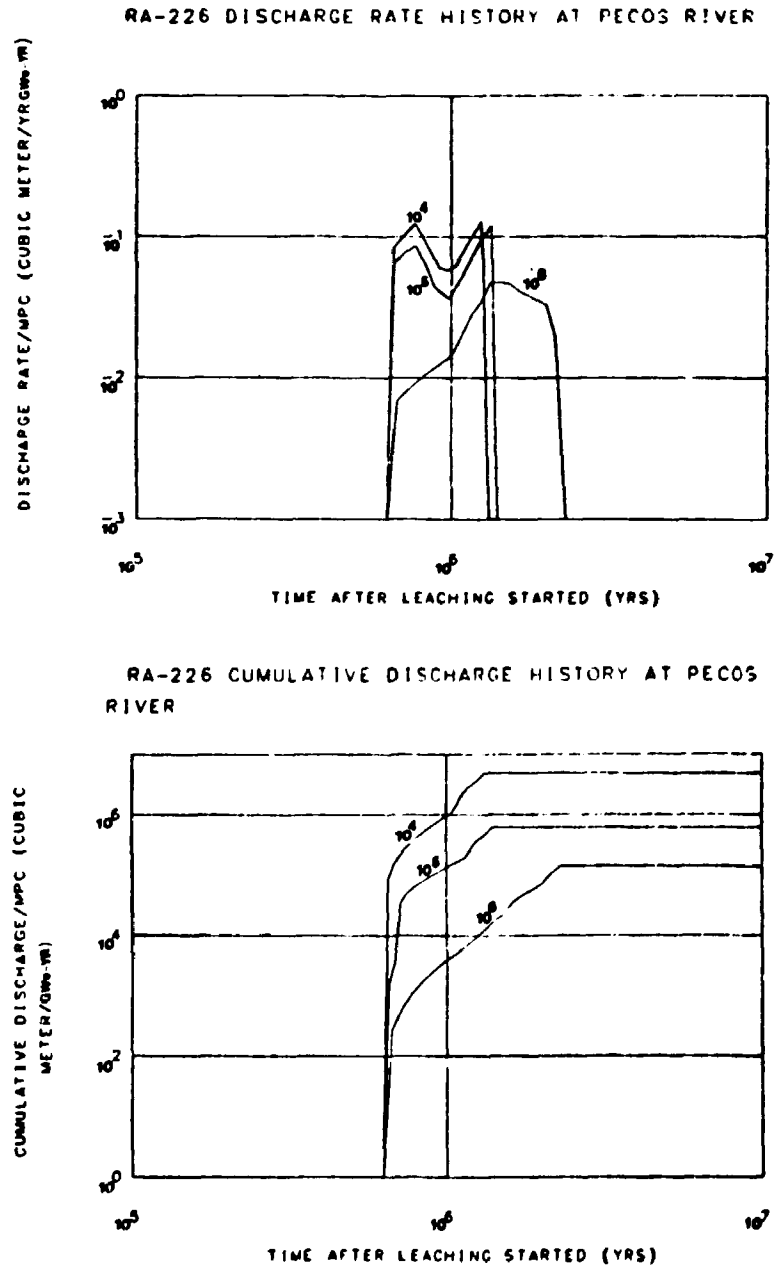


Figure 5 - Ra-226 discharge rate/MPC (water dilution rate) and cumulative discharge/MPC dilution volume at Pecos River discharging from Rustler aquifer (calculated data) for leach times of 10^4 , 10^5 and 10^6 years.

Comparable calculations were repeated for the Rustler aquifer, but in this case with the raw set of piezometric data. Using the raw set of piezometric data, the water dilution rates for I-129 and Ra-226 are comparable to those obtained by using the calculated potential data. However, I-129 reaches Pecos River after 10^4 years instead of $3.0E+4$ as in the previous case and Ra-226 reaches the river after $4.0E+5$ rather than $9.0E+5$ years.

During the period between 1937 and 1975 the maximum recorded Pecos River flow rate at Malaga Bend was $1.1E+11$ m³/yr while the lowest was $4.5E+6$ m³/yr. In the same period the average river flow rate was $1.7E+8$ m³/yr.

These Pecos River flow rates at the Malaga Bend are compared to the maximum total water dilution rate of 500 m³/yr for radionuclides discharging into Pecos River at any time. This maximum occurs for I-129, using the calculated potential and a leach time of 10^4 years. One concludes that when Pecos River is flowing at its minimum capacity it is still larger by a factor of 1000. Taking into account the fact that the amount of waste considered in the calculations is the result of production of one Gw(e)yr by a LWR, the Pecos River minimum flow rate would be sufficient to dilute to MPC levels the contamination caused by the reprocessed high level waste of 330 LWR's operating during their 30 years lifetime at full capacity.

At the WIPP site the downward seepage of contaminated groundwater into the underlying Delaware Mountain Group aquifers will be very unlikely. This is because the temperature gradient caused by the decay heat in the waste packages will produce an upward driving force due to density gradients. In addition, even after sufficient time for the heat generation to be significantly reduced, the hydraulic potential in the Delaware aquifer, being larger than in the Rustler aquifer, will cause the flow through any connection between them to be upwards.

Even if the Delaware aquifer were to be contaminated, the potential hazard due to the discharge of radionuclides into the Capitan aquifer situated about 30 km to the north would be small because the water travel times in this aquifer are large⁽³⁾.

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