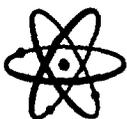
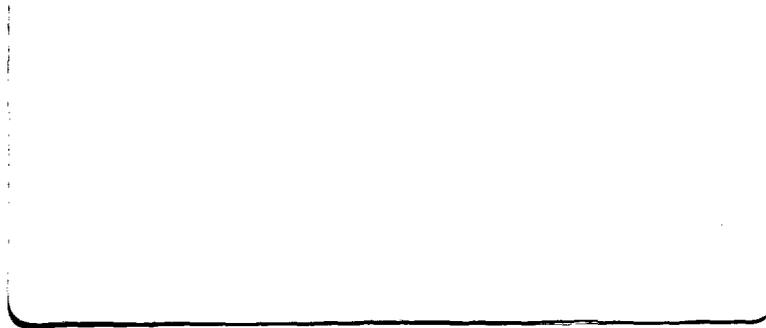


Encl. to June 24, 1987 ltr.
to Pohle from Logsdon



NUCLEAR WASTE CONSULTANTS INC.

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U.S. NUCLEAR REGULATORY COMMISSION
DIVISION OF WASTE MANAGEMENT

Technical Report #9

ANALYSIS OF THE IMPORTANCE OF
AQUITARD DIFFUSION DURING TRANSPORT BY
HORIZONTAL GROUND-WATER FLOW

Salt Repository Project
Subtask 3.5

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TECHNICAL ASSISTANCE IN HYDROGEOLOGY
PROJECT B - ANALYSIS
RS-NMS-85-009

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MINI-REPORT SUMMARY

Mini-Report Number: 9

Title: Analysis of the Importance of Aquitard Diffusion During Transport by Horizontal Ground-Water Flow

OBJECTIVE: The objective is to determine the importance of aquitard diffusion on the relative concentration of radionuclides within a zone of advection along a flow path from the waste repository to the accessible environment.

ANALYSIS: A steady-state analytical solution of coupled partial differential equations describing transport of radionuclides along an advective zone and diffusion of radionuclides into the bounding aquitard is used to estimate relative radionuclide concentrations in the advective zone at the accessible environment. Estimates of the relative concentrations are combined with estimated repository concentrations and simple darcy flow analyses to estimate the cumulative release to the accessible environment.

CONCLUSION: Relative concentrations of long-lived radionuclides are not significantly decreased by aquitard diffusion during advective transport to the accessible environment. Simple calculations suggest that EPA cumulative release limits may be exceeded for some radionuclides (e.g. Cs-135, Pu-240, Th-230).

DISCUSSION: The effects of adsorption of radionuclides was not considered in this analysis. Adsorption on the solid phase may provide a considerable sink for radionuclides and thus may be an important factor in radionuclide retardation. An additional analysis should be performed to investigate the effect of adsorption during advection.

ANALYSIS OF THE IMPORTANCE OF
AQUITARD DIFFUSION OF RADIONUCLIDES DURING TRANSPORT BY
HORIZONTAL GROUND-WATER FLOW

Numerical Evaluation of Conceptual Models
Subtask 3.5
Mini-performance Assessment #9

February 1987

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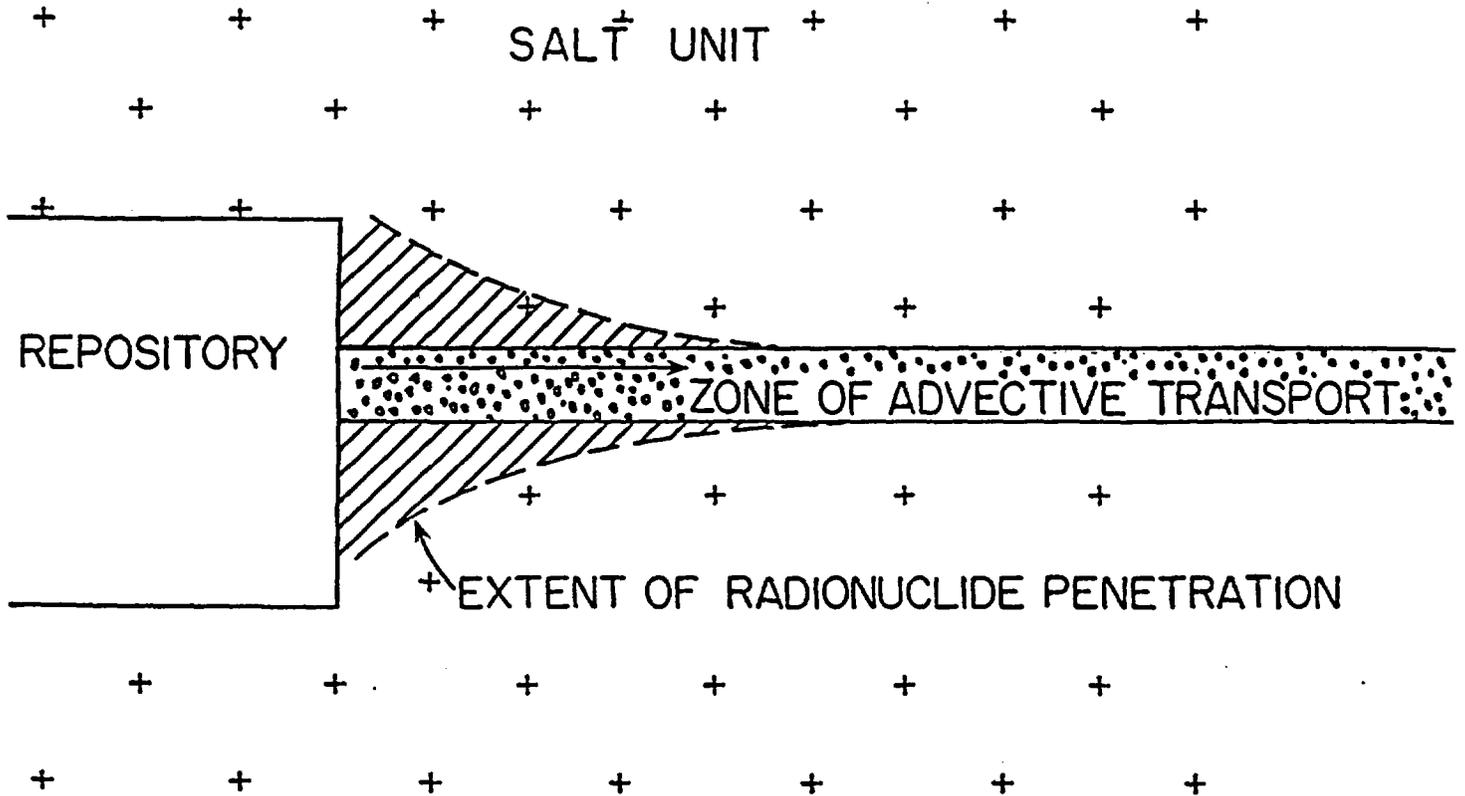
1.0 INTRODUCTION

1.1 General Statement of the Problem

Horizontal ground-water flow through interbeds within the Permian aquitard may provide a significant pathway for the transport of radionuclides to the accessible environment (Stephens & Assoc., 1986a). Diffusion of the radionuclides into the aquitard above and below the interbed may retard the transport of radionuclides to the accessible environment (Figure 1). The effect of aquitard diffusion depends on interbed thickness, magnitude of the dispersion coefficient and ground-water velocity (Sudicky and Frind, 1981).

1.2 Statement of Relevance to NRC

The location of the proposed repository is within the Permian evaporite aquitard in the Palo Duro Basin, Texas. Ground-water flow in the aquitard is generally considered to be downward, a favorable condition with regard to the ability of the repository to isolate waste (DOE, 1986; Stephens & Assoc., 1986b; 10CFR60, Section 60.122). Other studies, however, suggest that horizontal ground-water flow may occur within the Permian aquitard providing an alternate pathway of radionuclide transport to the accessible environment (for example, Stephens & Assoc., 1986a). The effect of aquitard diffusion during horizontal transport by ground-water flow through interbeds relates to the ability of the geologic repository to isolate waste and may



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Figure 1. Idealized Migration Pathway for Radionuclides Towards The Accessible Environment. During Transport Through The Zone of Advection (Interbed) Some of the Radionuclides Migrate into the Aquitard (Salt Unit) by Molecular Diffusion.

reduce the cumulative release of radionuclides to the accessible environment (see 10CFR60, Section 60.122).

1.3 Relationship to Other Analyses, Documents, Tasks and Subtasks

Most analyses consider hydrodynamic relationships within the Palo Duro Basin (Stephens & Assoc., 1986b); few, if any, analyses consider the process of aquitard diffusion of radionuclides within the Palo Duro Basin. Simple analyses suggest the possibility of significant horizontal ground-water flow through permeable units within the evaporite aquitard (see for example Senger and Richter, 1983; Stephens & Assoc., 1986a;). Results of one study (Stephens & Assoc., 1986a) suggest that permeabilities within the salt interbed, may be high enough that regulatory criteria may not be met. Therefore this analysis will examine the influence of aquitard diffusion on the transport of radionuclides during advection by flow through salt interbeds.

2.0 OBJECTIVE

The objective of the analysis is to determine the importance of aquitard diffusion by estimating the relative concentrations of radionuclides within a zone of advection as a function of distance along the flow path from the repository toward the accessible environment.

3.0 OPERATIONAL APPROACH - CONCEPTS AND GENERAL ASSUMPTIONS

3.1 Geology

The Palo Duro Basin has been described as being comprised of three hydrostratigraphic units (HSU); the upper fresh-water aquifer (HSU A) and the lower brine aquifer (HSU C) are separated by a Permian evaporite aquitard (HSU B, DOE, 1986; Figure 2). Typical rock types in the San Andres Formation (proposed location for a high level radioactive waste repository) include mudstone, limestone, dolomite, anhydrite, halite and terrigenous red beds (Kreitler and others, 1984; DOE, 1986).

The hydrologic system for this analysis is modelled as a layer of halite which contains a high level waste repository. Within the salt and intersecting the repository is a permeable, salt interbed which may provide a pathway for relatively rapid transport of radionuclides to the accessible environment by horizontal ground-water flow (Figure 1; Stephens & Assoc., 1986a).

3.2 Flow/Transport System

Ground-water flow is assumed to occur horizontally through the sedimentary interbed under saturated conditions. Transport of the radionuclides from the repository to the accessible environment occurs by advection. During transport through the interbed the migration of some of the radionuclides into the

ERA	SYSTEM	SERIES	GROUP	FORMATION	HYDROSTRATIGRAPHIC UNIT (HSU)
CENOZOIC	QUATERNARY			RECENT FLUVIAL AEOLIAN AND LACUSTRINE DEPOSITS	FRESHWATER FLOW SYSTEM HSU A
	TERTIARY			GCALLALA AND LACUSTRINE DEPOSITS	
MESOZOIC	CRETACEOUS	COMANCHE	WABITA		
			FREDRICKSBURG		
	TRIASIC		DOCKUM	TRUJILLO (Santo Rosa) TECOVAS	
PALEOZOIC	PERMIAN	OGCMA		DEWEY LAKE (Quaternary)	
				ALIBATES	
		GUADALUPE	ARTESIA		SALADO-TANSILL
				WHITEHORSE	YATES
			PEASE RIVER		SEVEN RIVERS
					QUEEN-GRAYBURG
		LEONARD	CLEAR FORK		SAM ANDRES (BLAINI)
					GLORIETA
					UPPER CLEAR FORK
					TUBS
					LOWER CLEAR FORK
					RED CAVE
				WICHITA	
				WOLFCAMP	
		PENNSYLVANIAN	VIRGIL	CISCO	
			MISSOURI	CANYON	
DES MOINES	STRAWN				
ATOKA	BEND				
MISSISSIPPIAN	MORROW				
	CHESTER				
	MERAMEC				
	OSAGE				
ORDOVICIAN	CANADIAN	ELLENBURGER			
CAMBRIAN		UNNAMED SANDSTONE			
PRECAMBRIAN					
				DEEP-BASIN FLOW SYSTEM HSU C	

Explanation

- Unconformity
- Boundary in Dispute

Figure 2. Generalized Hydrostratigraphic Column of The Palo Duro Basin (DOE, 1986).

aquitard occurs by molecular diffusion.

3.3 Repository and Source Term

The analysis will be performed using relative concentrations with the magnitude of the radionuclide relative concentration equal to unity at the repository.

4.0 TECHNICAL APPROACH

4.1 Formal Statement of Problem

Due to diffusive losses into the aquitard the transport of radionuclides to the accessible environment by horizontal groundwater flow through sedimentary interbeds will be retarded. The influence of these diffusive losses on the relative concentration within the zone of advection is examined.

4.2 Identification of solution Techniques

A steady-state analytical solution of coupled differential equations describing transport of radionuclides along the interbed and diffusion of radionuclides into the aquitard is presented by Sudicky and Frind (1981). Representative values for the aquitard/interbed system and a range of values for the radionuclide decay constant will be applied to the standard solution.

4.3 Definitions and Assumptions

The standard solution (Section 4.5) is described with the following terms:

- C/C_0 - steady-state, relative radionuclide concentration in the interbed
- V - interbed ground-water velocity
- D, D' - interbed, aquitard dispersion coefficient
- λ - radionuclide decay constant
- θ, θ' - aquifer, aquitard porosity
- $2b$ - interbed thickness

List of Assumptions:

1. Horizontal Darcy flow occurs through the zone of advection
2. Darcy flow in the salt is nonexistent
3. Salt dissolution does not occur
4. Concentration of radionuclide at repository is constant
5. System is at steady-state.
6. No adsorption of radionuclides

4.4 Identification of Expected Output and Judgement Criteria

The critical output of this analysis is the relative radionuclide concentration in the sedimentary interbed as a function of distance along the flow path. The relative radionuclide concentration in the zone of advective transport (the interbed) at the accessible environment (5 km, 40CFR191) will be the judgement criterion for this report. The significance of aquitard diffusion should be reflected by a decrease of

radionuclide concentration.

4.5 Standard Solution

The solution (Sudicky and Frind, 1981) used in this analysis is

$$c = c_0 \exp \left\{ \left[\frac{V}{2D} - \left(\left(\frac{V}{2D} \right)^2 + \frac{\lambda}{D} + \frac{\theta(AD)^{1/2}}{\theta b D} \right)^{1/2} \right] x \right\} \quad (1)$$

5.0 ANALYSIS

The parameter values listed below have been applied to the standard solution in order to obtain relative radionuclide concentrations within the zone of advective transport. Since these parameter values may not be representative of the aquitard interbed system, the analyses were performed for each parameter using a range of values for that individual parameter while keeping constant the values for the remaining parameters. The sensitivity of the solution to a particular parameter is thus illustrated.

D'	= 1.54 E -04 m ² /y
α_1	= 10 m
V	= 5.0 m/y
D	= $\alpha_1 V + D'$
λ	= values given in Table 1
θ'	= 0.01
θ	= 0.10
$2b$	= 1 m

RADIONUCLIDE	$t_{1/2}$ HALF-LIFE (years)	λ DECAY CONSTANT (1/years)
Americium-241	4.65×10^2	1.49×10^{-3}
-243	7.94×10^3	8.73×10^{-5}
Carbon-14	5.48×10^3	1.27×10^{-4}
Cesium-135	3.01×10^6	2.30×10^{-7}
-137	3.01×10^1	2.30×10^{-2}
Iodine-129	1.72×10^7	4.02×10^{-8}
Neptunium-237	2.19×10^6	3.16×10^{-7}
Plutonium-238	9.03×10^1	7.67×10^{-3}
-239	2.44×10^4	2.84×10^{-5}
-240	6.57×10^3	1.05×10^{-4}
-242	3.83×10^5	1.81×10^{-6}
Radium-226	1.62×10^3	4.29×10^{-4}
Strontium-90	2.74×10^1	2.54×10^{-2}
Technetium-99	2.11×10^5	3.29×10^{-6}
Thorium-230	7.94×10^4	8.73×10^{-6}
-232	1.40×10^{10}	4.96×10^{-11}
Uranium-233	1.62×10^5	4.29×10^{-6}
-234	2.49×10^5	2.78×10^{-6}
-235	7.12×10^8	9.74×10^{-10}
-236	2.38×10^7	2.91×10^{-8}
-238	4.38×10^9	1.58×10^{-10}

Table 1. Half-life and decay constant for several radionuclides. The half-life for each radionuclide is the time required for half of the radionuclides present to decay. The half-life for each radionuclide in this table is from Morgan (1977). The rate of decay of a radioactive isotope is proportional to the number of atoms of the radionuclide present. The constant of proportionality, the decay constant, is obtained by dividing the natural log of 2 by the half-life of the radionuclide (for example, Turcotte and Schubert, 1982).

6.0 RESULTS

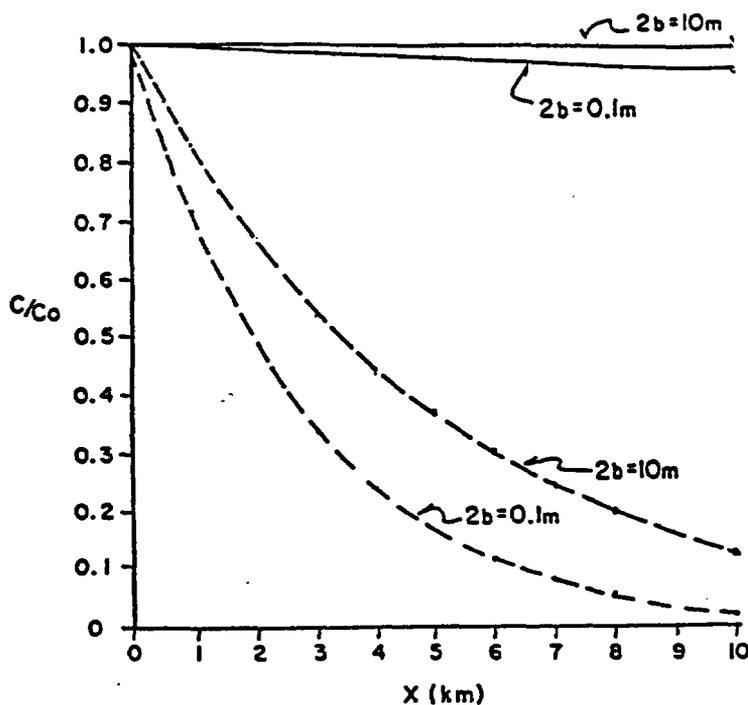
The numerical results are presented in graphical form in Figures 3 to 7. Figure 3 illustrates the significance of interbed thickness on the relative concentration along the ground-water pathway from the repository to the accessible environment. Similarly, the significance of interbed and aquitard porosity, the diffusion coefficient, the decay constant, and ground-water velocity are illustrated in Figures 4, 5, 6, and 7, respectively.

The topics of decay constant and ground-water velocity may be considered separate topics from the subject of this report, which is to evaluate the significance of diffusion. However, since these parameters are incorporated in the standard solution a brief discussion of these topics is included below to reflect this influence on the significance of diffusion.

6.1 Effect of Interbed Thickness

The effect of interbed thickness on the relative radionuclide concentration is shown in Figure 3. Mudstone interbeds are quite numerous within the lower San Andres Unit 4 Host horizon; the average thickness of the mudstone beds are 0.02 m with maximum thicknesses 0.5 m (DOE, 1986). Interbedded dolomites, anhydrites, and limestones are also present within the host horizon (DOE, 1986). Relative radionuclide concentration within

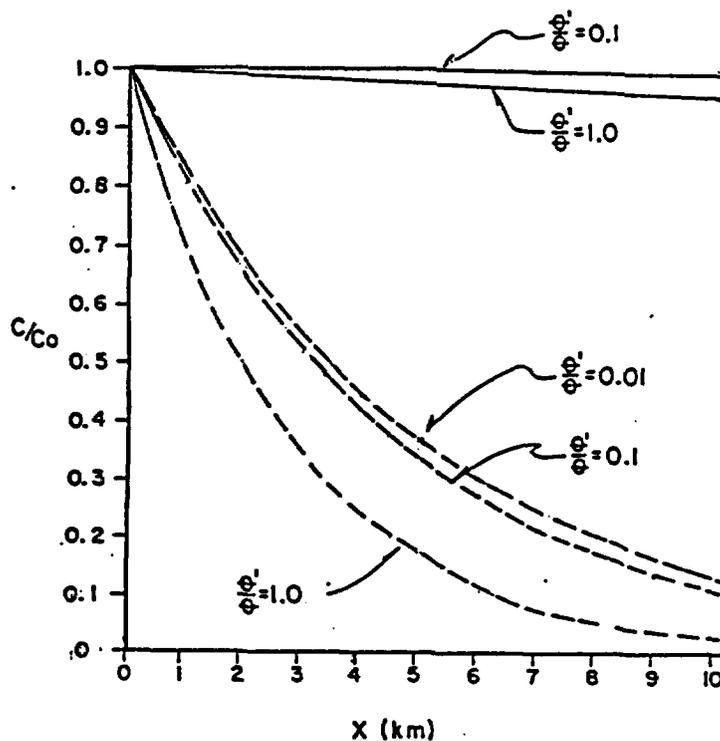
$v = 5 \text{ m/y}$
 $D' = 0.000154 \text{ m}^2/\text{y}$
 $\alpha = 10 \text{ m}$
 $\theta = 0.10$
 $\theta' = 0.01$
 - - - - $\lambda = 10^{-3} \text{ y}^{-1}$
 _____ $\lambda = 10^{-6} \text{ y}^{-1}$



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Figure 3. Relative radionuclide concentration (C/C_0) within the zone of advective transport as a function of distance (x) from the repository to the accessible environment. (See standard solution). Curves are for various values of the thickness of the zone of advective transport ($2b$). Values for the remaining parameters in the standard solution are given in the figure.

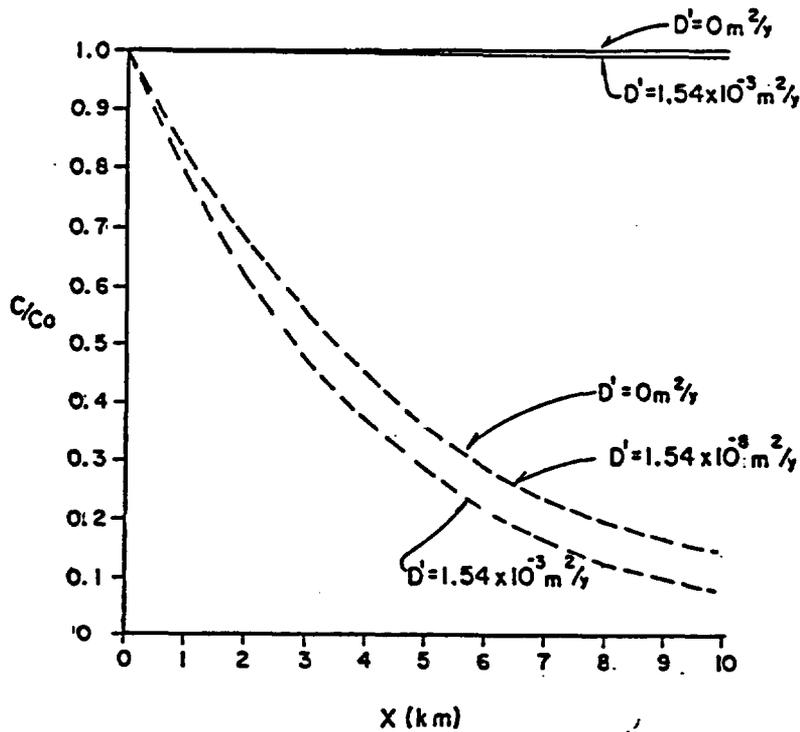
$v = 5 \text{ m/y}$
 $D' = 0.000154 \text{ m}^2/\text{y}$
 $\alpha = 10 \text{ m}$
 $2b = 1 \text{ m}$
 - - - - $\lambda = 10^{-3} \text{ y}^{-1}$
 _____ $\lambda = 10^{-6} \text{ y}^{-1}$



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Figure 4. Relative radionuclide concentration (C/C_0) within the zone of advective transport as a function of distance (x) from the repository to the accessible environment. (See standard solution). Curves are for various values of the aquitard porosity relative to the porosity within the zone of advective transport (θ'/θ). Values for the remaining parameters in the standard solution are given in the figure.

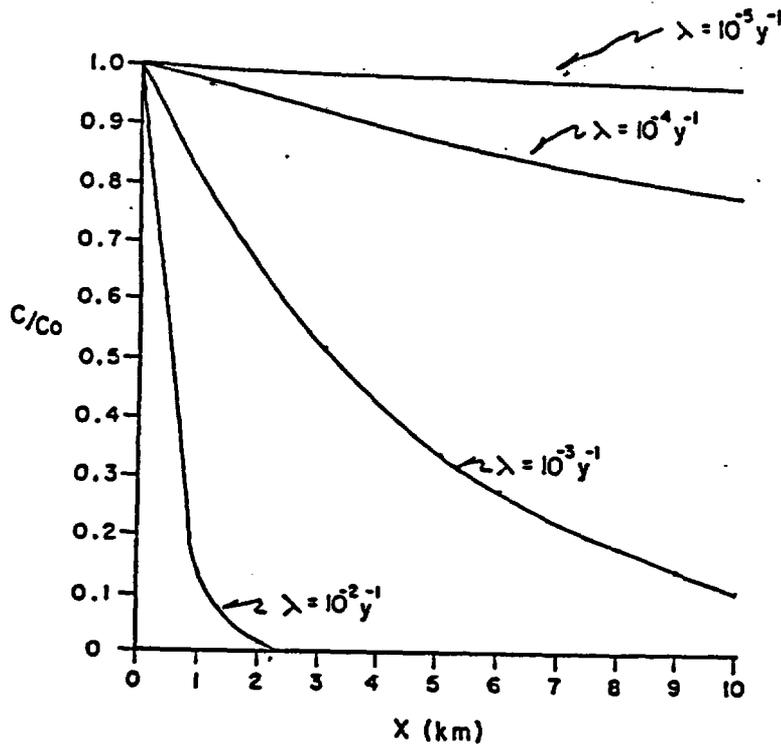
$v = 5 \text{ m/y}$
 $\theta = 0.10$
 $\theta' = 0.01$
 $2b = 1 \text{ m}$
 - - - - $\lambda = 10^{-3} \text{ y}^{-1}$
 _____ $\lambda = 10^{-6} \text{ y}^{-1}$



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Figure 5. Relative radionuclide concentration (C/C_0) within the zone of advective transport as a function of distance (x) from the repository to the accessible environment. (See standard solution). Curves are for various values of the diffusion coefficient (D'). Values for the remaining parameters in the standard solution are given in the figure.

$v = 5 \text{ m/y}$
 $D' = 0.000154 \text{ m}^2/\text{y}$
 $\alpha = 10 \text{ m}$
 $\theta = 0.10$
 $\theta' = 0.01$
 $2b = 1 \text{ m}$



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Figure 6. Relative radionuclide concentration (C/C_0) within the zone of advective transport as a function of distance (x) from the repository to the accessible environment. (See standard solution). Curves are for various values of the decay constant (λ). Values for the remaining parameters in the standard solution are given in the figure.

$$D' = 0.000154 \text{ m}^2/\text{y}$$

$$\alpha = 10 \text{ m}$$

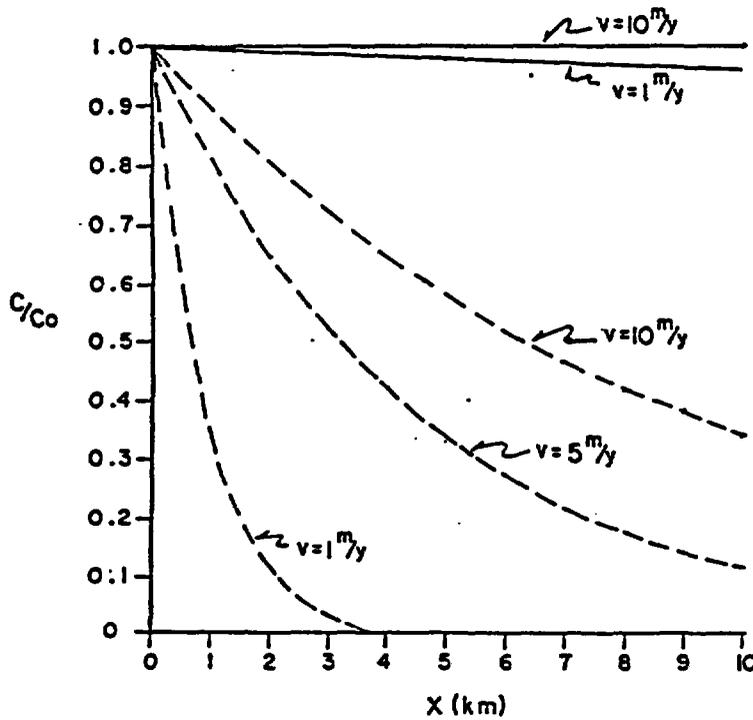
$$\theta = 0.10$$

$$\theta' = 0.01$$

$$2b = 1 \text{ m}$$

----- $\lambda = 10^{-3} \text{ y}^{-1}$

_____ $\lambda = 10^{-6} \text{ y}^{-1}$



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Figure 7. Relative radionuclide concentration (C/Co) within the zone of advective transport as a function of distance (x) from the repository to the accessible environment. (See standard solution). Curves are for various values of the ground-water velocity in the zone of advective transport (v). Values for the remaining parameters in the standard solution are given in the figure.

the interbeds will decrease as the interbed thickness decreases (Figure 3). The significance of interbed thickness decreases as the radionuclide decay constant decreases (Figure 3).

6.2 Effect of Porosity

The results presented in Figure 3 were obtained using interbed and aquitard porosities of 0.10 and 0.01, respectively. The DOE (1986, Tables 3-14 to 3-15) reports neutron porosities for the lower San Andres Formation (J. Friemel No. 1 well) ranging from less than 0.01 to greater than 0.10. Aquitard diffusion is, however, essentially a function of the magnitude of the aquitard porosity relative to the interbed porosity (see standard solution, Figure 4). Figure 4 includes relative concentration profiles for the case where the interbed and aquitard porosities are equal. A decrease in the interbed porosity relative to the aquitard porosity results in a decrease in the relative radionuclide concentration within the zone of advection (Figure 4). The importance of the difference in porosities between the interbed and aquitard is diminished for radionuclides with relatively small decay constants (see Figure 4).

6.3 Effect of Diffusion Coefficient

The profiles presented in Figure 5 illustrate the difference in relative radionuclide concentration expected for two cases where the aquitard diffusion coefficients differ by two orders of

magnitude. The DOE (1986) reports an empirical brine diffusion coefficient of $1.5 \times 10^{-4} \text{ m}^2/\text{y}$. This empirical brine diffusion coefficient has been used to describe the transport of radionuclides (dissolved in the brine) via brine diffusion (DOE, 1986, p. 6-261). Brine migration away from the waste package results from a concentration gradient of brine in the solid salt. The values for the diffusion coefficient used to obtain the profiles in Figure 5 are an order of magnitude larger and smaller than the empirically derived brine diffusion coefficient reported by the DOE (1986, p. 6-261, 262). The DOE (1986) also states that this empirically derived brine diffusion coefficient is about 4 orders of magnitude larger than the solid state diffusion coefficient which would apply if the radionuclides were to diffuse through the salt by themselves. In any case, the magnitude of the diffusion coefficient seems to have limited significance on the relative radionuclide concentration for relatively small radionuclide decay constants (Figure 5).

6.4 Effect of Decay Constant

Relative concentration profiles along the zone of advective transport are presented in Figure 6 for several different values of the decay constant. Decay of radionuclides in the aquitard will affect the concentration gradient at the aquitard boundary and therefore also influence the diffusion of radionuclides into the aquitard (see Appendix).

The relative concentration of radionuclides at a distance of 5 km from the repository is significantly diminished (compared to the concentration at the repository) for radionuclides with decay constants greater than about 10^{-4} y^{-1} (Figure 6). Many radionuclides, however, have decay constants less than 10^{-5} y^{-1} (Table 1).

6.5 Effect of Velocity

Transport of radionuclides by horizontal ground-water flow through interbeds of the Evaporite Aquitard is a separate issue from aquitard diffusion and has been addressed in a previous study (Stephens & Assoc., 1986a). The significance of aquitard diffusion on radionuclide concentration is, however, influenced by the magnitude of ground-water velocity in the zone of advective transport; increasing the ground-water velocity diminishes the effect of aquitard diffusion (see standard solution, Figure 7). Figure 7 illustrates the relative concentration within the interbed for several values of ground-water velocity; these values have been approximated giving consideration to regulatory criteria (i.e. 5 km in 1000 or 10,000 years). Dutton and Orr (1985) estimate horizontal specific discharge in the carbonate beds of the San Andres formation to be about $3 \times 10^{-4} \text{ m/y}$ ($8.8 \times 10^{-7} \text{ m/day}$). Assuming a value of 0.01 for porosity suggests a ground-water velocity of 0.03 m/y. For ground-water velocities much less than 0.5 m/y (5km in 10,000 years), aquitard

diffusion should not be a significant issue in terms of cumulative release criteria. Another study (Stephens & Assoc., 1986a), however, suggests (based on the limited amount of permeability data) that cumulative release criteria (40CFR191) may not be met.

Consider now numerical estimates of the maximum cumulative release of radionuclides to the accessible environment which include the effects of aquitard diffusion. The cumulative release of radionuclides to the accessible environment (CR_{AE}) is defined here as the cumulative release of radionuclides from the repository (CR_R) multiplied by the relative concentration (C/C_0 , see standard solution) at the accessible environment (5km):

$$CR_{AE} = CR_R \left(\frac{C}{C_0}\right)_{x=5km} \quad (2)$$

where

$$CR_R = Q \cdot t \cdot C_0 \quad (3)$$

and Q is the total volume flux of water through the repository, t is effective release time and C_0 is the concentration of nuclides at the repository.

An estimate of the flux of water through the repository is obtained from estimates of average ground-water velocity, porosity and cross-sectional area of the repository (5 m/y, 0.10, and 1000 m², respectively; velocity and porosity are discussed earlier in the text, repository cross-sectional area estimated from a general description given in the Final Environmental

Assessment DOE, 1986, p. 5-25).

The effective release time is the total time that ground-water (with a constant concentration of radionuclides defined by the standard solution) passes from the controlled zone into the accessible environment (5 km down gradient from the repository). For an average ground-water velocity of 5 m/y radionuclides released from the repository at the time of emplacement will not be released to the accessible environment (5 km) until about 1000 years (ground-water travel time) after emplacement. Thus ground water contaminated with radionuclides will enter the accessible environment for only 9,000 years during the 10,000 years following waste emplacement. Therefore, the effective release time is estimated to be 9,000 years.

A worst-case estimate of the concentrations of the individual radionuclides is obtained from solubility limits presented by the DOE (Table 6-33, p. 6-232 to 6-236; 1986). The relative concentration of the radionuclides at the accessible environment has been calculated with the standard solution using the parameter values given at the top of Figure 3 (interbed thickness $2b = 1\text{m}$) and the appropriate decay constant from Table 1.

The estimates of the maximum cumulative release of several radionuclides to the accessible environment are compared in Table 2 with EPA limit, (40CFR191). The estimates exceed the

Radionuclide	(1) Solubility (g/m ³)	CR _P Cummulative Release From Repository (g)	(2) C/C _o at Accessible Environment	CR _{AE} Cummulative Release at Accessible Environment (g)	Activity (Ci)	EPA Limit Assuming 70,000 MTIM in Repository (Ci) (Assuming 1,000 MTIM)
Americium-241 -243	0.0001	450	0.21 0.90	95 405	302 75	700 (100) *exceeded
Carbon-14	0.06	270,000	0.86	232,200	1.09 x 10 ⁶	700 * (100) *
Cesium-135 -137	600,000.0	2.7 x 10 ¹²	0.999 <10 ⁻⁸	2.7 x 10 ¹² <27,000	2.4 x 10 ⁹ <2.3 x 10 ⁶	7,000 * (1,000)*
Iodine-129	600,000.0	2.7 x 10 ¹²	0.999	2.7 x 10 ¹²	4.4 x 10 ⁸	700 * (100) *
Neptunium-237	0.001	4,500	0.998	4,491	3	700 (100)
Plutonium-238 -239 -240 -242	0.001	4,500	0.00042 0.96 0.88 0.995	1.9 4,320 3,960 4,478	32 266 896 17	700 * (100) *
Strontium-90	0.8	3,600,000	<10 ⁻⁸	<0.04	<5	7,000 (1,000)
Technetium-99	0.001	4,500	0.992	4,464	77	70,000 (10,000)
Thorium-230 -232	0.001	4,500	0.98 0.9999	4,410 4,500	87 4.5 x 10 ⁻⁴	70 * (10) *
Uranium-233 -234 -235 -236 -238	0.001	4,500	0.991 0.993 0.9999 0.9995 0.9999	4,460 4,469 4,500 4,498 4,500	43 27 9.7 x 10 ⁻³ 0.3 1.6 x 10 ⁻³	700 (100)

(1) DOE, 1986, Table 6-33

(2) Standard Solution.

* = exceeded

Table 2. Estimate of Maximum Cumulative Release of Radionuclides

limits for the following radionuclides (assuming 70,000 MTHM in repository; see Table 2):

- Carbon-14
- Cesium-135
- Plutonium-240
- Thorium-230

7.0 CONCLUSIONS

Analyses have been performed to estimate the relative concentration of radionuclides within a zone of advection as a function of distance along the flow path from the waste repository to the accessible environment. The decay constant of the radionuclides is a key parameter in the standard solution for these analyses. While many of the aquitard and interbed parameters may profoundly affect the relative concentration of some of the radionuclides (for example Americium-241, Carbon-14, and Strontium-90, Table 1), these hydraulic parameters have little effect on the concentration of radionuclides with relatively small decay constants (for example, Uranium-235, Thorium-232, and Plutonium-242, Table 1). Simple calculations incorporating aquitard diffusion suggest that cumulative release limits may be exceeded for several radionuclides including Carbon-14, Cesium-135, Plutonium-240 and Thorium-230. Therefore, it is concluded, within the limits of the assumptions made in this report, that aquitard diffusion will not be a significant process

in the ability of the geologic repository to isolate the waste.

8.0 DISCUSSION

A previous study (Stephens & Assoc., 1986a) has demonstrated the possibility of ground-water flow through interbeds within the Evaporite Aquitard. Based on a very simplified analysis, it was concluded that permeabilities within the aquitard may be sufficient for ground-water travel time and cumulative release criteria to be exceeded. The study did not consider the importance of aquitard diffusion of radionuclides which would decrease the amount of radionuclides being transported towards the accessible environment.

The significance of aquitard diffusion (and radioactive decay) on the concentration of radionuclides in a zone of advective transport has been investigated in this report. The results of this study suggest that the decay constant of the radionuclides is a key parameter affecting the cumulative release of radionuclides to the accessible environment. Aquitard diffusion and radioactive decay may significantly diminish the concentration of radionuclides with relatively high decay constants (for example Carbon-14, see Sudicky and Frind, 1981). However, the concentration of radionuclides with relatively low decay constants (for example, Uranium-235) is not greatly reduced by aquitard diffusion (given the assumptions made in this

report). Other hydrogeologic parameters used to characterize the aquifer/aquitard system have little effect on the cumulative release of many of the radionuclides expected to be found in the repository (Table 1).

This report has not considered the effects of adsorption of radionuclides. Adsorption on the solid phase in the aquitard may provide a considerable sink for radionuclides and thus may be an important factor in radionuclide retardation (see Neretnieks, 1980). The solubility of the radionuclides will determine to some extent the cumulative release of radionuclides to the accessible environment. Sensitivity analyses to examine the significance of possible ranges of solubility have not been performed in this report.

It is suggested that aquitard diffusion should not be considered as a significant, favorable geochemical condition affecting the ability of the repository to isolate the following radionuclides: C-14, Cs-135, Pu-240, Th-230. Potential zones of advective transport which are in proximity to the proposed repository location should be identified during site characterization because the effect of these zones on the ability of the repository to isolate the four radionuclides listed above will not be significantly compensated for by aquitard diffusion and radioactive decay. An additional analysis should be performed to investigate the significance of radionuclide adsorption during transport by horizontal ground-water flow.

9.0 REFERENCES

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APPENDIX

APPENDIX

The following derivation is taken from Sudicky and Frind (1981).

The radionuclide concentration in a very long aquifer/aquitard system at steady-state can be described by the following differential equation.

$$D \frac{d^2c}{dx^2} - V \frac{dc}{dx} - \lambda c = \frac{q}{\theta b} \quad 0 \leq x \leq \infty \quad (1)$$

where "c" is the radionuclide concentration, "x" is distance along the aquifer, "D" is the dispersion coefficient, "V" is the ground-water velocity in the aquifer, " λ " is the decay constant of the radionuclide, "q" is the diffusive radionuclide flux into the aquitard, " θ " is the aquifer porosity and "2b" is the aquifer thickness. The boundary conditions for equation 1 are

$$\begin{aligned} c(0) &= c_0 \\ c(\infty) &= 0 \end{aligned} \quad (2)$$

The differential equation describing molecular diffusion of radionuclides in the aquitard is

$$D' \frac{d^2c'}{dx^2} - \lambda' c' = 0 \quad b \leq x \leq \infty \quad (3)$$

where "D'" is the diffusion coefficient, "c'" is the radionuclide concentration in the aquitard. The boundary conditions for the thick aquitards are

$$\begin{aligned} c'(b, x) &= c(x) \\ c'(\infty, x) &= 0 \end{aligned} \quad (4)$$

where "z=b" is the contact between the aquifer and aquitard. The diffusive flux of radionuclides is (Fick's Law)

$$q = -\theta D' \frac{\partial c'}{\partial z} \Big|_{z=b} \quad (5)$$

where " θ " is the aquitard porosity.

The diffusion coefficients (D') for the aquitard and aquifer, although probably not actually equal, are assumed to be equal in order to simplify the solution. The dispersion coefficient " D "

$$D = \alpha_L V + D' \quad (6)$$

results from molecular diffusion and mechanical mixing. The parameter " α_L " in equation 6 is the longitudinal dispersivity.

The radionuclide concentration in the aquitard is

$$c' = c \exp \left\{ -(\lambda/D')^{1/2} (z - b) \right\} \quad (7)$$

Equation 7 was obtained by solving Equation 3 assuming continuity of radionuclide concentration at the aquitard/aquifer interface. The concentration gradient (see equation 5) is obtained by differentiating equation 7 with respect to z. The radionuclide concentration gradient at the aquitard boundary is

$$\frac{dc'}{dz} \Big|_{z=b} = -c \left(\frac{\lambda}{D'} \right)^{1/2} \quad (8)$$

Therefore the aquifer losses due to aquitard diffusion (equation 5) are given by

$$q = \theta'D'c \left(\frac{\lambda}{D}\right)^{\frac{1}{2}} \quad (9)$$

which may be substituted into equation 1.

A solution to equations 1 and 2 is

$$c = c_0 \exp \left\{ \left[\frac{V}{2D} - \left(\left(\frac{V}{2D} \right)^2 + \frac{\lambda}{D} + \frac{\theta'(\lambda D)^{1/2}}{\theta b D} \right)^{1/2} \right] x \right\} \quad (10)$$

which is the standard solution in Section 4.5 of this report.

A simple FORTRAN program to calculate the standard solution follows. The program has been checked by hand calculations.

```

c      aquitard diffusion model, sudicky and frind
c      initialize parameters
double precision a, f, rnum, den, e, d
double precision arg, coefx, c
real lmda
write(5,*)' enter velocity, m/y'
read(5,*)v
write(5,*)' enter aquitard diffusion coefficient, m2/y'
read(5,*)dp
write(5,*)' enter longitudinal dispersivity, m'
read(5,*)alpha
write(5,*)' enter decay constant, 1/y'
read(5,*)lmda
write(5,*)' enter aquifer porosity'
read(5,*)theta
write(5,*)' enter aquitard porosity'
read(5,*)thetap
write(5,*)' enter aquifer thickness, 2b, m'
read(5,*)b
b=b/2.
write(5,*)' enter distance to accessible environment, km'
read(5,*)dist
dist=dist

d=alpha*v+dp
write(5,*)' v= ',v,' m/y'
write(5,*)' dprime= ',dp,' m2/y'
write(5,*)' alpha= ',alpha,' m'
write(5,*)' d= ',d,' m2/y'
write(5,*)' lmda= ',lmda,' 1/y'
write(5,*)' theta= ',theta
write(5,*)' thetaprime= ',thetap
write(5,*)' dist= ',dist,' km'
c      combine parameters
a=v/(2*d)
f=lmda/d
rnum=thetap*((lmda*dp)**.5)
den=theta*b*d
e=(a**2.)+f+rnum/den
e=e**.5
write(5,*)' a=',a
write(5,*)' e=',e
write(5,*)' d=',d
write(5,*)' f=',f
write(5,*)' rnum=',rnum
write(5,*)' den=',den
coefx=a-e
write(5,*)' coefx=',coefx
write(5,*)'      x(km)
c'

```

```
c      apply variable to solution and print results
do 1 i=1,11
      r=(float(i)-1.)*dist/10.
      rl=r
      arg=coefx*r*1000.
      c=dexp(arg)
      write(5,2)rl,c
      format(1x,2x,f8.2,5x,f10.8)
2
1      continue
      stop
      end
```