# PERFORMANCE OF MONOLITHIC CONCRETE WASTE FORMS

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## Abstract

Liquid wastes can be made into concrete or cement waste form that can be poured into a concrete vault forming a monolith. The waste isolation performance of monolithic concrete waste forms or vaults is generally dominated by the influence of cracks through the structure. In relation to water flow rate and crack spacing, monolithic concrete vaults have three general regions of performance. At extremely low flow rates, release is strictly diffusionally limited. In most situations, flow rates will not be low enough to ensure diffusional release. At slightly greater flow rates (the magnitude of which is dependent upon the diffusion coefficients and crack spacing), release is controlled by the flow rate of water through cracks in the structure with release rate approximately proportional to Darcy flow. In this region, release is not sensitive to block size and the vault behaves as an equivalent porous medium from a mass transport perspective. At higher flow rates, release rate is controlled by diffusion out of intact blocks of waste form. In this situation the release rate is very sensitive to block size (crack spacing) but independent of flow.

## INTRODUCTION

One option for disposal of liquid radioactive or hazardous wastes is to form a concrete or cement waste form which can be poured into a concrete vault forming a monolith. Monolithic waste forms have many advantages including structural integrity, longevity, and relative ease of emplacement. In order to design monolithic concrete vaults and concrete waste forms for high waste isolation performance it is necessary to better understand the factors controlling performance.

The single greatest weakness of concrete from the perspective of radioactive waste isolation is the tendency to crack. Cracks create preferential pathways in otherwise impermeable systems, leading to enhanced leaching of contaminants. This paper examines the role of cracking and flow rate on waste isolation performance of monolithic concrete vaults including suggestions for improved vault design.

Concrete is a brittle material with high compressive strength but low tensile strength. These characteristics make massive concrete structures prone to cracking. Although concrete degradation is typically modeled as loss of effective thickness of the concrete (1,2), in fact the most widespread type of concrete degradation is extensive cracking. Examination of old railroad bridges reveals that frequently the original surface of the concrete is essentially intact, while the body of the structure is riddled with cracks. The bridges are earth covered on the top and open on the bottoms. Essentially all of the cracks show evidence of water flow in the form of carbonated material on the edges of the cracks and active dripping of water from the cracks.

New concrete structures also are prone to cracking as a result of temperature induced volume changes and shrinkage as can be observed visually on most massive concrete structures. Drying shrinkage is affected by many factors which include unit water content, aggregate composition, and duration of initial moist curing (USDI, 1988). Initial drying shrinkage ranges from less than $2x10^{-4}$ for dry, lean mixes with good quality aggregate to over $10^{-3}$ for rich mortars or concretes containing poor quality aggregate (3). Autogenous volume change related to chemical reactions and aging of the concrete may also cause shrinkage in the range of $10^{-5}$ to $1.5x10^{-4}$ (3).

Temperature induced volume changes occur primarily from the heat of hydration which causes expansion during early time periods while the concrete has higher creep. When the concrete cools it then shrinks, leading to cracking. The cooling occurs at a later stage, after the concrete has aged and relief of stress by creep is lower.

## MODELING APPROACH

The concrete vault is envisioned as a large fractured monolith, with blocks of intact concrete separated by fractures. Water percolates through the fractures while transport in the matrix is by diffusion. A number of semi-analytical solutions have been developed for transport through fractured porous media in the literature. Of available methods, the solution of Rasmuson and Neretnieks (1981) is perhaps most appropriate for application to release rate from massive concrete vaults. A decaying source term is assumed which is required for leaching and the solution is given in terms of dimensionless parameters which can be used interpret and generalize the results from the analysis. The published solution estimates transport from a decaying source of radionuclides into a fractured porous media. The solution for leaching from unsaturated waste forms is the complement of the case for release into a porous medium.

The parametric study is limited to the case of no dispersion in the fractures in the concrete. This is the worst or conservative case for radionuclide release rate from concrete waste forms.The no dispersion case is not always conservative for the case of radionuclide transport in groundwater as is assumed by many analysts. Fractures in concrete may be subject to channelization of flow in the fractures. This, as well as variability in fracture spacing and location, would effectively make the concrete monolith behave as some unknown combination of the situations evaluated in this work.

The fracture flow solution assumes that the only transport in the z direction is inside the fractures (i.e., advection in the fractures is much greater than diffusion through the matrix). At very low water flow rates this assumption will not be true and transport in the z direction will be dominated by diffusion. One dimensional diffusion out of the monolith can be estimated with an error function solution. Parameter space where pure diffusion could denominate is shown by the shading in the graphics.

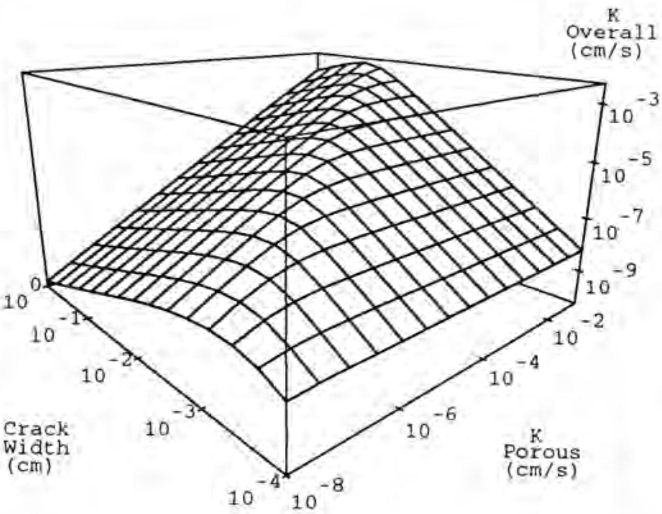

Fig. 1. Relationship between crack width, hydraulic conductivity of adjacent porous material, and effective hydraulic conductivity of vault roof.

The statement is sometimes made that cracks will not influence the performance of concrete vaults located in the unsaturated zone because water held under tension will not enter the cracks. There are two major problems with this presumption. First, massive vaults tend to promote formation of perched water on the vault roof which can migrate directly into the cracks (5). Second, any cracks are likely to become partially filled with porous material, allowing imbibition of water under tension into portions of the crack.

Mass transport through concrete vaults depends heavily upon water flow rates. Usually the mass transport will occur out the bottom of the vault whereas water flow rate is controlled by the vault roof. The leakage rate through the roof is dependent upon water supply, crack spacing in the roof, and the permeability of the porous material near the roof. If a low permeability porous material such as clay is placed next to the roof, flow rates through the vault can be expected to be in the range of $10^{-8}$ cm/s or below throughout most of the vault *lifetime*. Figure 1 illustrates the results for a crack fraction of $10^{-4}$. If the cracks are partially sealed with water stops, then the hydraulic conductivity will be even lower. Conversely, if higher permeability materials such as sand or gravel are placed next to the vault, then the effective hydraulic conductivity in the presence of cracks can be very high.

Transport of contaminants in the concrete matrix is predominantly by diffusion. Although diffusion is conceptually simple, estimation of diffusion rates in concrete can be complicated and error prone. The major problems are lack of standardization of nomenclature and the necessity of lumping several poorly understood processes into the diffusion coefficient. When diffusion experiments are performed the rate of flux of a particular ion is measured. This rate of flux may depend upon many factors including:

a. the tortuosity and constrictivity of the porous medium (i.e., concrete waste form),
b. adsorption of the ion onto the solid phase,
c. precipitation/dissolution of the ion as a solid,
d. solid solution of the ion in components of the concrete,
e. complex formation and speciation in solution,
f. electrical potential gradients related to differential ion diffusion rates,
g. physical entrapment of the ion in the concrete, and
h. radioactive decay.

Subsequent analysis of the experimental data generally results in some or all of the above processes being lumped into the resultant diffusion coefficient. Depending upon how the experiment was performed, and the type of calculations used in data analysis, the reported diffusion coefficient can mean many different things. For this reason, great care must be used when applying published diffusion coefficients for concrete in performance assessment calculations. Consistency *must* be maintained between the experimental methodology and subsequent use of experimentally determined diffusion coefficients.

Groundwater codes which consider transient contaminant migration in heterogeneous media are written in terms of a diffusion coefficient which generally does not include sorption, porosity, entrapment of the contaminant, or solubility limits subsumed inside the diffusion coefficient. The distinction becomes critical in heterogenous systems because the direction and rate of diffusion is actually controlled by concentration gradients in the fluid phase, not gradients in total concentration. Unless great care is taken to rectify measured diffusion coefficients which represent several parameters with

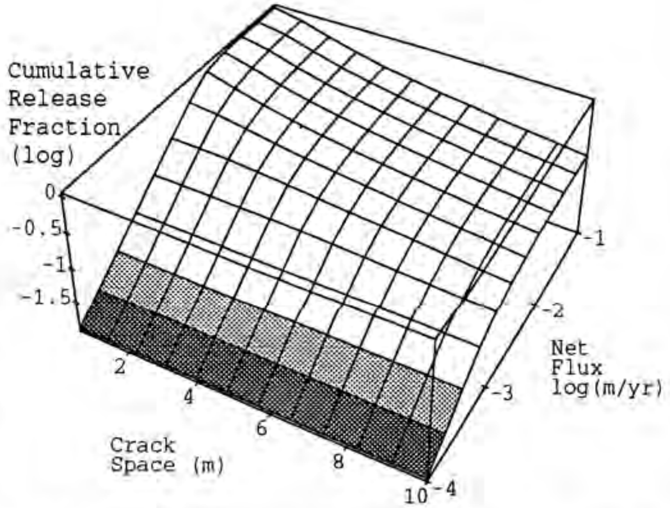

Fig. 2. Cumulative release fraction of nitrate over 500 years as a function of crack spacing and water flux through vault.

the diffusion coefficient required in groundwater codes, gross errors in the performance assessment will occur.

## CUMULATIVE RELEASE CALCULATIONS

One of the performance measures applicable to any waste disposal system is the cumulative release of contaminants over a fixed time period. If the linear dose-response hypothesis for exposure to radioactivity is correct then the total health impact of the site will be approximately proportional to total release of radionuclides. Cumulative releases are convenient because this eliminates time as a variable to plot, allowing illustration of a wide range of parameter values on a single figure.

In the calculations four nominal contaminants are considered: nitrate, technetium, chromium, and tritium. Nitrate is considered only because there is a substantial amount of data on nitrate leaching. Nitrate behavior is synonymous with long-lived radionuclides which have high solubility in concrete waste forms and little adsorption onto the solid phase (e.g., iodine, oxidized technetium). Technetium and chromium are subject to adsorption and solubility limitations in some mixes. Tritium is not subject to solubility limitations or adsorption and has a short half life of 12.7 years.

Data for diffusion coefficients are taken from Serne and Wood (6). The values used in the figures are $D_a = 5\times10^{-8}$ $cm^2/s$ for nitrate, $5\times10^{-8}$ $cm^2/s$ for tritium, $10^{-8}$ $cm^2/s$ for technetium, and $10^{-10}$ $cm^2/s$ for chromium. The vault is assumed to be 10 meters thick and the waste form porosity is 0.4.

Cumulative release in Fig. 2 to Fig. 5 is given as the logarithm to the base 10 of the fraction of the initial inventory which is released. A value of 0 indicates total release of the initial inventory. For non-decaying contaminants such as nitrate, eventually all of the inventory will be released in any scenario. For decaying contaminants, delays in release reduce the total release rate (i.e., much decays prior to potential release). The graphs are intended to illustrate controls on release rate and do not correspond to any actual disposal system currently being built.

The region of possible diffusional dominance of release and where flow becomes unimportant is illustrated by shading. This transition zone to pure diffusional control is dependent upon the material placed around the vault. In most situations, a layer or shell of concrete or other materials will be placed around the concrete waste form, greatly lowering pure diffusional release rates below the values given in the shaded regions.

Three general regions are apparent in each of the graphs. The location of each region is dependent upon the radionuclide of concern, crack spacing, the properties of the waste form, and the amount of water percolation into the vault. At higher water percolation rates the release rate becomes independent of water flow rate, but is highly dependent upon fracture spacing. This upper region represents the case where the contaminant concentration in the fractures is essentially zero. Release rate is controlled by diffusion from the interiors of the blocks to the cracks which is highly dependent upon crack spacing. Once the water percolation rate is rapid enough to hold the concentration in the cracks to near zero, additional water flow has little effect on release rate. The second region occurs at slightly lower flow rates and is characterized by the release rate being independent of crack spacing but highly dependent upon water percolation rate. In this region the contaminant concentrations in the cracks and matrix are essentially the same and the system behaves as an equivalent porous medium.

The third region occurs at very low water percolation rates and is characterized by a constant release rate. At very low flow rates, the release rate is entirely diffusionally controlled and neither flow rate nor crack spacing are important in influencing release rate.

The location of each performance region in the parameter space of crack spacing and water flow rate (Darcy velocity through vault) is dependent upon the apparent diffusion coefficient and radioactive decay. Contaminants which are bound to the solid such as chromium (low apparent diffusion coefficients) are released at much lower rates, but the release rate is sensitive to flow rate over a much broader range. From a mass transport perspective, it is not always important to be able to estimate crack spacing and flow rate in order to determine the performance of a monolithic concrete vault. In

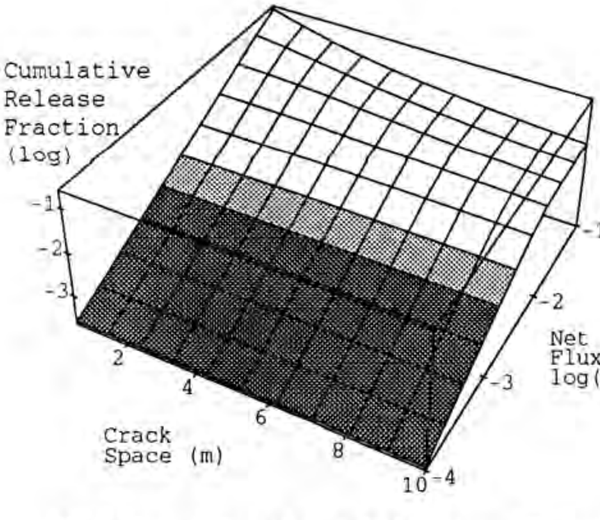

Fig. 3. Cumulative release of tritium over 500 years.

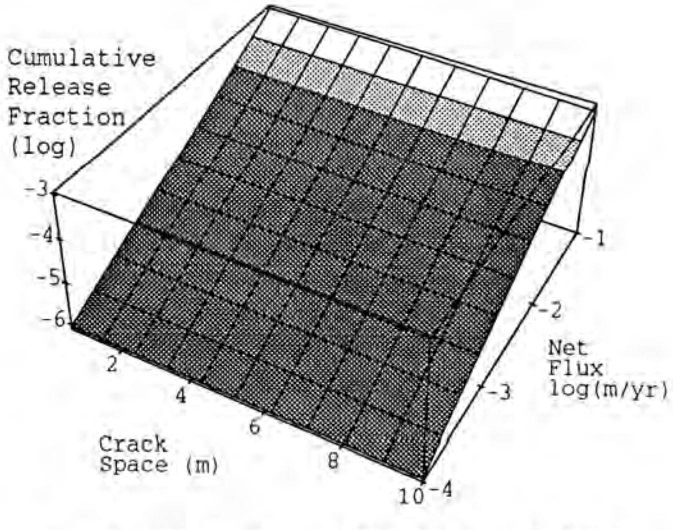

Fig. 4. Cumulative release of chromium over 500 years.

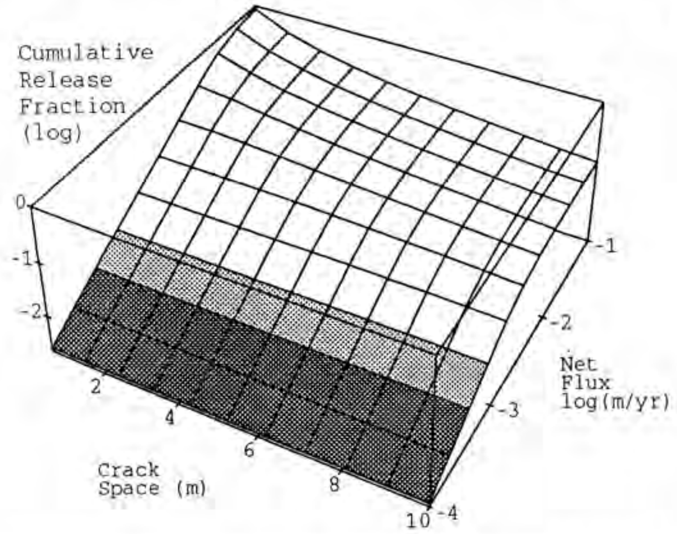

Fig. 5. Cumulative release of technetium-99 over 500 years.

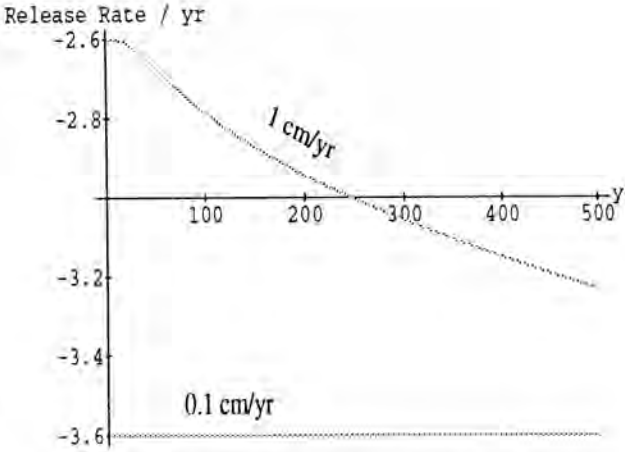

Fig. 6. Release rate of nitrate from a simulated concrete monolith with changing water percolation rate. Release rate is the logarithm to the base 10 of the fractional release rate.

many instances, only one of the two key parameters will be important. Unfortunately, the important parameters may be different for each contaminant being leached.

## CONCENTRATION VS. RELEASE

The previous section evaluated the controls on release rate of contaminants from a concrete vault. In general, the population exposure and therefore (assuming no threshold for adverse effects) total excess cancers are directly proportional to total contaminant release. In contrast, the maximum dose to an exposed individual is related to maximum concentration in groundwater. The behavior of concrete vaults in terms of concentration in the effluent is frequently different than total release or release rate. At low water percolation rates through the vault, the cracks maintain the same concentration as the pore water in the matrix. At more rapid flow rates, the excess water passing through the cracks does not increase release but does provide dilution water.

The calculations for nitrate illustrate the phenomena. Figure 6 gives the logarithm of the fractional release rate as a function of time for water percolation rates of 1 cm/yr and 0.1 cm/yr. The release rate is much greater at the higher flow rate.Figure 7 illustrates the relative concentration of the effluent for several different water flow rates. The relative concentration is the concentration in the liquid exiting the vault normalized to the total concentration initially placed in the grout waste form (i.e., solid + liquid concentration per unit total volume porous media). Because nitrate is all in the pore fluid, the maximum relative concentration is simply the inverse of the porosity ($1/0.4 = 2.5$). In the figure, the concentration of the effluent is much higher for the low flow, low release situation.

The crossover effect between release rate and effluent concentration behavior has interesting implications for performance assessment. Since the performance standards for low level waste and most hazardous waste are based (indirectly) upon concentrations in the groundwater and not on total release rate, increasing the water flow rate through the vault (e.g., by failure of the engineered cover) can actually facilitate compliance with regulatory standards. This is another example where a clearly conservative assumption for performance assessment calculation (i.e., early cover failure) can turn out to be non-conservative and lead to an underestimate of dose rates.

Although the calculations indicate that concentrations in the effluent coming out of the vault will increase at low flow rates, concentration based standards are typically enforced in the groundwater some distance from the vaults (e.g., at the site boundary). Thus the effluent will have an opportunity to mix with groundwater and the final concentration will be dependent upon both release rate and effluent concentration. Depending upon parameters such as dispersion and depth of the aquifer, either release rate or effluent concentration could dominate downstream well concentrations.

## DESIGN IMPLICATIONS

The general behavior of groundwater concentrations has implications for vault design. Most disposal facilities for low level radioactive waste and hazardous wastes will consist of a series of vaults or trenches. The engineered cover can either be designed to cover the entire facility or each individual vault/trench. A single cover over the entire facility will tend to increase the proportion of water which goes into surface runoff rather than subsurface recharge. Additionally the large cover places any recharge water away from the vaults. Multiple, smaller covers over each vault/trench (lower portion of Fig. 8) may be more expensive but will provide dilution water for the leachate without increasing contaminant release rate from the vault.

Another design feature to improve performance is to begin the engineered cover design at the vault, rather than at the ground surface. Most covers are designed in terms of functional layers (e.g., plant growth layer, lateral drainage, resistance, capillary break, etc.) which begin at the earth's surface. The space between the bottom of the cover and the vault is filled with available "backfill" soils. An alternative is to begin construction of the cover at the surface of the concrete vault. The space between the top of the cover and the surface grading can be filled with backfill.

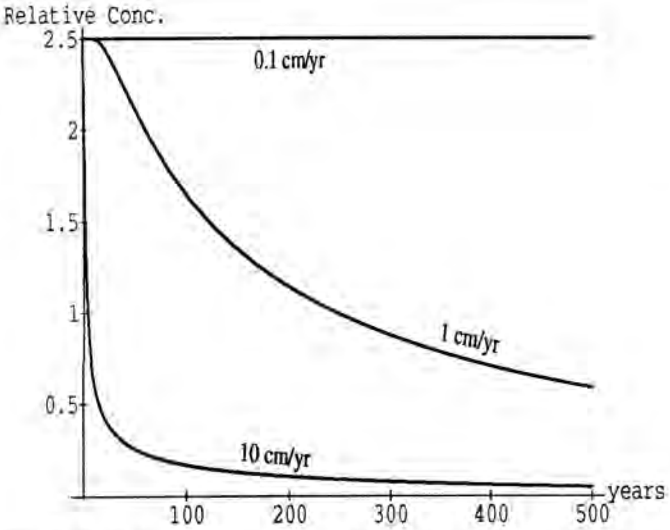

Fig. 7. Relative concentration of nitrate in effluent for different flow rates.

The alternate design places the clay layer adjacent to the concrete vault. Placing the clay layer against the vault will lower flow rates through cracks in the concrete (5) and slow degradation of the concrete. Placing the layers of the cover at greater depths and against the concrete provides greater protection against cover disruption by subsidence, plant roots, burrowing animals, and erosion.

From a standpoint of compliance with regulatory standards, the multiple small covers are generally preferable to a single large cover. This conclusion is true not only for monolithic concrete vaults but also for traditional shallow trench disposal of radioactive waste. The effect of dilution water is an example of where sometimes a humid climate location may theoretically be advantageous over an arid site (at least from the narrow viewpoint of regulatory compliance).

As with any generalized statement concerning waste isolation performance, there are exceptions. In cases where the vadose zone is very deep, a large cover over the entire disposal site will provide longer travel times to the groundwater. If the travel time is significant, relative to radionuclide decay rate, and relative to the containment period offered by the vault, then a large cover may provide superior performance. This is potentially the case for very arid sites.

## CONCLUSION

Monolithic concrete vaults have three general regions of performance. At extremely low flow rates, release is strictly diffusionally limited. In most situations, flow rates will not be low enough to ensure diffusional release.

At slightly greater flow rates (the magnitude of which is dependent upon the diffusion coefficients), release is controlled by the flow rate of water through cracks in the structure with release rate approximately proportional to Darcy flow. In this region, release is not sensitive to block size and the vault behaves as an equivalent porous medium from a mass transport perspective.

At higher flow rates, release rate is controlled by diffusion out of intact blocks of waste form. In this situation the release rate is very sensitive to block size (crack spacing) but independent of flow.

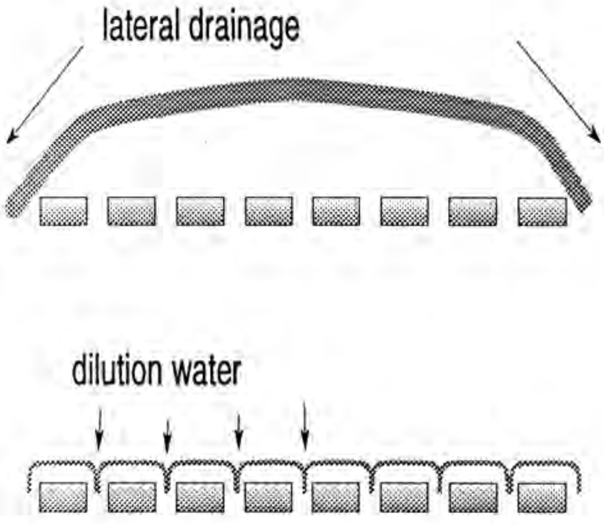

Fig. 8. Two alternative designs for an engineered cover over a series of concrete vaults.

Calculations also indicate that increased water flow rate around and through the vaults in not always negative from a maximum exposed individual viewpoint. Greater water flow rates through concrete vaults may actually improve performance relative to regulatory standards by lowering maximum concentrations in groundwater.

## ACKNOWLEDGMENT

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