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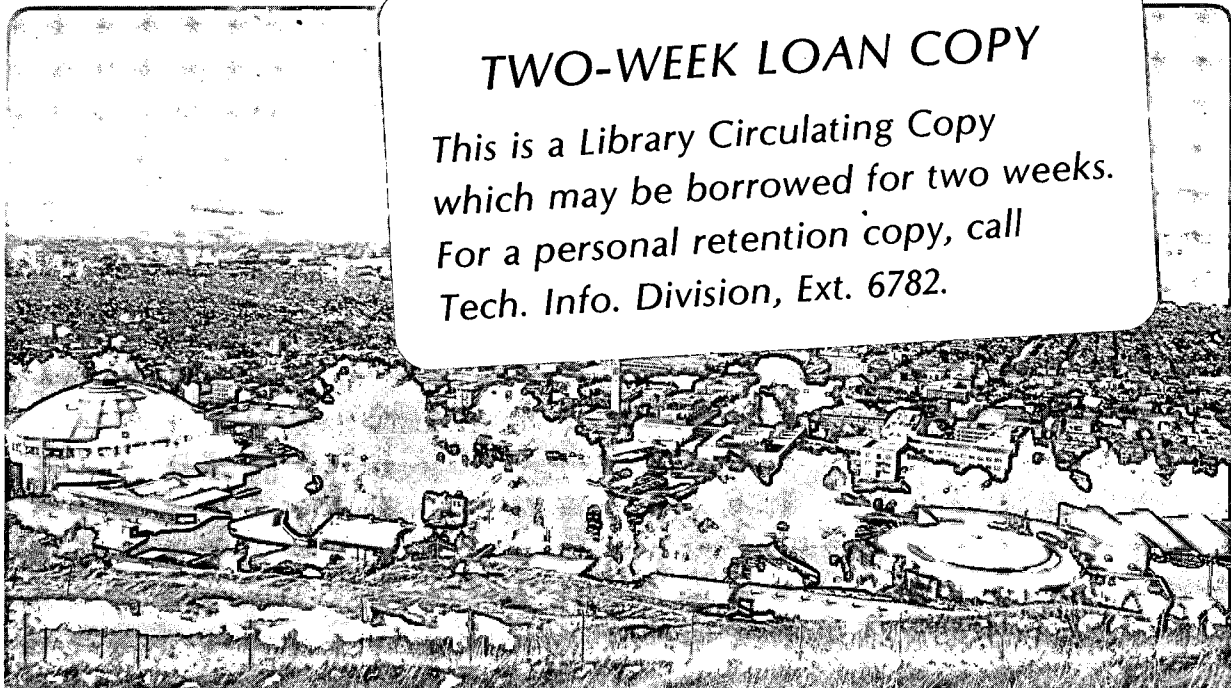
GEOLOGIC DISPOSAL OF RADIOACTIVE WASTE - 1983

T.H. Pigford

October 1983

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GEOLOGIC DISPOSAL OF RADIOACTIVE WASTE - 1983

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Presented at "Nuclear Chemical Engineering - Then, Now, and in the Future ----- as viewed by Robert E. Wilson Awardees", American Institute of Chemical Engineers (AIChE) 75th Anniversary Meeting, Washington, D.C., November 2, 1983.

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Abstract

Geologic repositories for radioactive waste are evolving from conceptualization to the development of specific designs. Estimates of long-term hazards must be based upon quantitative predictions of environmental releases over time periods of hundreds of thousands of years and longer. This paper summarizes new techniques for predicting the long-term performance of repositories, it presents estimates of future environmental releases and radiation doses that may result for conceptual repositories in various geologic media, and it compares these predictions with an individual dose criterion of 10^{-4} Sv/yr.

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The 1981 Robert E. Wilson Lecture [1] on geologic disposal dealt with the long-term potential for contaminating water with waste radionuclides. It presented some simple quantitative estimates of the relative long-term hazards from buried radioactive wastes, from surface piles of uranium mill tailings, and from surface piles of coal ash containing uranium and its decay products. These estimates were based on assumed dissolution rates of solid wastes and assumed properties for hydrogeologic transport.

New techniques have since been developed to predict the rates of dissolution in a geologic environment and to predict the time-dependent transport of radionuclide chains through geologic media. Data on typical hydrologic and geochemical properties of various geologic media and estimates of radiation doses resulting from released radionuclides have been evaluated by the Waste Isolation System Panel, of the National Research Council, which has recently completed a three-year evaluation of the long-term performance of the geologic isolation system for radioactive waste [2]. These new predictive techniques, their use with the new system data, and the resulting estimates of the long-term hazards from geologic isolation [2,3] are summarized herein. Needs for further data and analysis are identified.

The estimates of long-term hazards, presented here as radiation doses to maximally exposed individuals, are compared to the individual dose criterion of 10^{-4} Sv/yr for expected events, adopted in the National Research Council study. The form of the performance criterion, whether the radiation dose to the maximally exposed individual or, as recently proposed by EPA [4], the maximum amount of radioactivity released over a fixed period of 10,000 years, can materially affect the view of what constituents adequate long-term isolation.

Radionuclide inventories in conceptual geologic repositories

Conceptual repositories are assumed here to contain radioactive waste resulting from reprocessing 10^5 Mg of uranium fuel from light-water reactors, with radionuclide inventories given in Table 1. These inventories include radionuclides in high-level and transuranic wastes, as well as separated carbon-14 and iodine-129.

Waste dissolution - limitations of present leaching experiments

In previous analyses [1,5,6,7] it has been assumed that waste solids dissolve congruently, i.e., the fractional dissolution rate of each radioactive constituent in the waste solid is assumed to be the same as that of the waste matrix, and dissolution rates have been inferred from laboratory leaching experiments that measure the rate of chemical reaction between waste solids and simulated groundwater. However, most of the radionuclides in waste do not dissolve congruently [2]. Many of

the important radioelements are so insoluble that they cannot go into solution as rapidly as the waste matrix itself. Such solubility effects are exhibited in laboratory leaching experiments with borosilicate glass waste and with other waste solids, but these experiments do not simulate the mass-transfer limitations on dissolution that would occur in a geologic repository.

Dissolution controlled by solubility-limited bulk flow

Wood et al. [8,9] treat solubility-limited transport as originating with a volume flow of groundwater uniformly saturated with each solubility-limited radionuclide as the groundwater flows across the plane of waste emplacement. This is an unrealistic and unnecessarily conservative assumption in that it neglects the important resistance in the concentration boundary layer adjacent to the waste surface, and it assumes instantaneous diffusive and dispersive transport laterally across the flow cross section. Although useful for upper-bound estimates of solubility-limited transport, such estimates do not reflect a valid physical model of waste dissolution and transport.

Dissolution limited by mass transfer

The analyses by Chambre' et al. [10,11,12] predict the rates at which radionuclides in liquid at the surface of a waste package can be transported into groundwater in the surrounding porous medium. The calculation assumes that the concentration of each elemental species in the water at the surface of the waste package is at a maximum value given by the solubility of that species. This assumption was suggested from the laboratory experiments by Rai and Strickert [13], who observed that the equilibrium concentrations of low-solubility actinides in static leachant in contact with borosilicate glass were equal to the solubilities of the stable compounds of those species in the leachant, and by the many observations [14,15,16] of actinides and other low-solubility constituents in precipitates formed on borosilicate-glass surfaces during laboratory leaching experiments.

It is assumed that the waste form is in contact with porous rock or with backfill of the same porosity and permeability as the rock. The profiles of groundwater flow, of the concentration of a dissolved radioelement, and of the resulting plume of contaminated water flowing past the waste form are illustrated in Figure 1. Diffusion and convection of the individual dissolved constituents through this concentration boundary layer are analyzed to predict the limiting dissolution rate of low-solubility radionuclides. Effects of gaps and different backfill properties are discussed elsewhere [2,17].

Dissolution rate controlled by diffusion and convection -- The diffusion-and-flow calculation [10,12] makes use of the known distribution of ground water velocities around a semi-infinite

cylinder through the pores of the surrounding rock. Assuming Darcy flow, the velocity distribution is mathematically equivalent to the "potential flow" of theoretical hydrodynamics. The rate of diffusion through such a fluid is assumed to be represented by Fick's Law, using a coefficient for molecular diffusion in the liquid in the rock pores. In the present estimates we assume that the diffusion coefficient is equal to that in a free liquid, conservatively disregarding the tortuosity of the pores.

The fractional release rate f_j of the elemental species j , and of its isotopes, is calculated at steady state to be:

$$f_j = \frac{8\epsilon N_j^* (D_j U)^{1/2} (1+R/L)}{(\pi R)^{3/2}}, \quad \frac{UR}{D_j} \quad 4 \quad (1)$$

where N_j^* is the concentration in the groundwater at the waste surface, D_j is the liquid diffusion coefficient, L is the length of the waste cylinder, R is its diameter, U is the pore velocity of the ground water before it comes near the waste, ϵ is the porosity of the surrounding rock, and n_j is the bulk density (g/cm^3) of elemental species j in the waste. To predict the maximum solubility-limited dissolution rate, N_j^* is the solubility.

Table 2 gives values of the water solubility of silica and of some radioelements important in long-term geologic isolation. Also listed are the bulk densities and the calculated fractional release rates for a typical borosilicate glass waste, exposed to groundwater at an approach velocity of 1 m/yr in a rock of one percent equivalent porosity. The assumed diffusion coefficient of $10^{-5} \text{ cm}^2/\text{sec}$ is typical for an electrolyte in water. It conservatively neglects the effect of tortuosity [18], which in granite can result in more than a 100-fold [19] to 1000-fold [20] reduction in D , and can result in a 10- to 30-fold reduction in the fractional dissolution rates estimated from Equation (1).

Dissolution controlled by liquid diffusion -- When the conditions are such that UR/D_j is much less than four, Equation (1) must be replaced by an equation [11,12] for the steady-state release from a waste package embedded in a porous medium saturated with stagnant water ($U = 0$):

$$f_j = \frac{\beta \epsilon D_j N_j^*}{n_j}, \quad U = 0 \quad (2)$$

where β is a geometrical factor. For a sphere of radius R :

$$\beta = \frac{3}{R^2} \quad (3)$$

For a prolate spheroid approximating the cylindrical waste form:

$$\beta = \frac{3e}{b^2 \ln(\coth \frac{\alpha_s}{2})} \quad (4)$$

where b is the semi-minor axis, e is the eccentricity, and α_s is a parameter that defines the spheroid surface:

$$\alpha_s \equiv \cosh^{-1}(\frac{1}{e}) \quad (5)$$

A cylindrical waste form of dimensions given in Table 2 can be approximated by $\beta = 37 \text{ m}^{-2}$.

Using the properties listed in Table 2, the limiting zero-velocity fractional release rate for SiO_2 , at 20°C and steady state, is calculated to be $8.7 \times 10^{-7}/\text{yr}$, about one fourth the release rate calculated at $1 \text{ m}/\text{yr}$. The much lower diffusion coefficients inferred from experiments in granite [19,20] could result in 100- to 1000-fold reductions in the predicted diffusion-controlled dissolution rates.

Comparison with laboratory leach data -- Table 2 gives values of the fractional release rates for silica and for radioactive elements, calculated from reported experimental values. For these species, the values of f_i computed from Equation [1] are smaller than several values derived from the laboratory leach tests. One must conclude, therefore, that when waste is embedded in repository rock and is surrounded by slowly moving groundwater the slow diffusion and slow movement of the liquid around the waste containers can be more significant in controlling dissolution than the rate at which the substances inside the waste material reach the surface of the waste form. If the solubility is small and/or the inventory of the elemental species in the waste large, the rate of escape into ground water will be determined primarily by the properties of the rock and the velocity of ground water, although the chemical environment and solubilities near the waste surface may be affected by the waste material and by radiolysis. If the solubility is sufficiently large the kinetics of the interaction between the solid waste constituents and water may dominate.

Application to repositories -- Predicted solubility-limited fractional dissolution rates in Table 2 for the low solubility species are much smaller than the fractional release rates estimated from laboratory leaching experiments wherein the concentration boundary layer resistance is either negligible or absent. This seems true for borosilicate glass waste and for other waste solids, such as Synroc. Thus, for a waste form in the repository environment, surrounded by ground water in a porous rock, the rate of dissolution for these low-solubility

species will be controlled by the rate of transport into the liquid boundary layer adjacent to the waste solid. For most radionuclides, except cesium, the kinetics of the solid-liquid interactions are rapid enough that the concentration of the dissolved species in the liquid at the waste surface will be near or only a little below the solubility limits for these species. Because the kinetics of the solid-liquid interactions are not controlling for these low-solubility species, the laboratory leach rate data are irrelevant and are not useful for estimating the dissolution of low solubility species under repository conditions.

From analyses for the long-term environmental releases for wastes from fuel reprocessing, the soluble cesium-135 is the only radionuclide important to long-term repository performance whose dissolution rate from glass may not be affected by solubility-limited dissolution, if cesium can dissolve faster than the waste matrix itself. If the hazard from Cs-135 is not affected by its rate of dissolution, as is indicated later, then the rate of dissolution or degradation of borosilicate glass itself would not affect repository performance.

The diffusion-convection theory is used later in this paper to calculate source terms and dissolution rates of high level, transuranic, and other wastes assumed to be emplaced in the conceptual repositories.

Dependence on waste form material properties -- If the chemical species of radionuclides released to groundwater are the same as assumed in the calculations for Table 2, then the fractional dissolution rates of Table 2 would apply to any other waste form of the same surface area and waste loading, unless the waste matrix itself is so insoluble that its fractional release rate is less than that calculated for the solubility-limited radionuclides. This does not occur for borosilicate glass, but it is possible for waste forms more insoluble than any of those now tested.

In a repository environment the solid-liquid interactions in borosilicate glass are rapid enough for low-solubility species to be at or near their solubility limits in the adjacent liquid, so interior cracks in the waste solid, devitrification, and other such mechanisms that could increase the rate of the solid-liquid interaction are not expected to affect the solubility-limited dissolution rates.

The chemical and material properties of the waste form itself can affect the formation of stable precipitate layers and colloids, which can affect the applicability of the solubility-limited diffusion-convection analysis.

Effects of solubility uncertainties and colloids --- The above analyses of the solubility-limited dissolution rates are useful for predicting the long-term release of radionuclides in a

repository if the solubilities are known and if there are no other mechanisms that can release and transmit radioactivity to the biosphere. The solubilities of many of the species are sensitively affected by the local oxidizing-reducing conditions of the ground water and the host rock, the acidity or alkalinity, the temperature, and the presence of complexing species such as anions and some organics. Some indications of these effects on solubility are given in Table 3. The solubilities must be known for such analyses to be used as a basis for repository design.

The rate of release of low-solubility species to ground water can be increased if colloids and suspended precipitates form at or near the waste surface. It may be possible for such suspended matter to be transported to the biosphere by groundwater as it flows through the porous or fractured media, in addition to the material transported in solution. If colloidal particles form they may transport at the water velocity if the particles are smaller than the rock pores, and if they are not sorbed [24]. Otherwise, colloidal particles may be trapped in the pores and not move [25,26].

Conversely, if the reported solubilities include an appreciable amount of species such as colloids, there will be less in solution and there will be a lower rate of diffusive transport into the concentration boundary layer than estimated in Table 2. The effective diffusion coefficients of colloids are very low, about $5 \times 10^{-7} \text{ cm}^2/\text{sec}$ for 5×10^{-6} -cm colloids in a liquid [27], even without tortuosity corrections. Therefore, because the diffusion coefficient of colloids is small compared with that of solutes, only the solute fraction of the "solubility" will diffuse appreciably into the groundwater.

The time to reach steady state; validation experiments -- Although the predictive techniques of Equations (1) and (2) contain no unknown constants to be empirically determined, the techniques do need validation by carefully designed laboratory experiments. In planning such experiments, the time to reach the steady-state conditions is important. The time-dependent, solubility-limited dissolution rates have been derived by Chambre [11,12]. For a non-sorbing specie whose decay during movement through the concentration boundary layer can be neglected, the "equilibration time" for the release rate to come within one percent of the steady-state diffusion-controlled rate of Equation (2) is 310 yr for the parameters given in Table 2. The time is much shorter, of the order of a few years, when the flow velocity is great enough to warrant use of Equation (1). For a sorbing specie the equilibration time is proportional to the specie's retardation coefficient in the rock. Although these equilibration times are short compared to the times of interest for storage, they are long enough to be important in designing experiments to test the predictions of these boundary-layer controlled rates.

The steady-state conditions of Equations (1) and (2) can

be reached within reasonable experimental times by choosing parameters different from those expected in a repository, and this is a reasonable approach for testing the predictive technique for mass transfer in liquids in porous media. Validation under simulated repository conditions may have to depend on careful tests of the time-dependent dissolution rates predicted from the more general theory that led to Equations (1) and (2).

Carbon-14 waste

Carbon-14 is not a constituent within the high-level waste packages, because it is separated in fuel reprocessing. However, if carbon-14 is recovered separately and emplaced in a geologic repository, we can estimate its solubility-limited dissolution rate. Reprocessing 10^5 Mg of uranium fuel will yield 19.8 kg of carbon-14 as CO_2 in the off gas, diluted with 1.18×10^5 kg of natural carbon [28]. It is assumed that the CO_2 is converted to granular calcium carbonate and loaded into 1100 waste canisters, each with dimensions given in Table 2. The heat generation rate within each carbon-14 package will be only a few milliwatts, so the carbon-14 packages can be at ambient temperature.

The solubility of calcium carbonate at 25°C is 1.4×10^{-5} g/cm³ [29]. If the carbon-14 waste package is of the same dimensions as the high-level waste package in Table 6, we estimate a solubility-limited fractional dissolution rate of carbon-14 of 4×10^{-7} /yr.

Iodine-129 waste

We assume that the iodine recovered in fuel reprocessing is to be emplaced in a geologic repository and that the iodine is converted to a suitably insoluble compound loaded into canisters of the same dimensions as assumed for high-level waste. Although no chemical or physical form of the iodine waste has been specified, we assume here that iodine is as silver iodide, with a density of 5.6 g/cm³. For an estimated solubility of the contained iodine of 1×10^{-8} g/cm³ [29], and correcting for dilution by the stable iodine-127 in the waste, the estimated solubility-limited dissolution rate of iodine is 8×10^{-11} /yr. The fractional dissolution rate of the silver iodide waste is then low enough that most of the iodine-129 will decay before it is released from the waste package.

The total mass of iodine in each waste package would be 560 kg, with a heat generation rate of a fraction of a watt, so there is no self heating. About 40 waste packages would contain the 22.4 Mg of total iodine recovered from the 10^5 Mg of uranium fuel, and 18.8 Mg of silver would be required. Assuming the May 1982 cost of industrial silver of 0.17 \$/g, the total cost of silver to combine with the iodine would be \$3.3 million. Lead iodide would be cheaper, but its greater solubility would result

in a dissolution rate of about 7×10^{-6} /yr. silver iodide waste has been assumed for this study.

Although we have assumed in this study that separated carbon-14 and iodine-129 is to be emplaced in a geologic repository, ocean disposal is an alternative to be considered.

Calculation of the transport of radionuclides to the environment

The long-term performance of a geologic repository for radioactive waste can be expressed as the lifetime dose commitment to future individuals who drink water contaminated by radionuclides from the repository and who eat food grown from the contaminated water. The adequacy of geologic disposal can be measured by comparing calculated future radiation doses from released radionuclides to some performance criterion. For expected events, such as long-term dissolution and hydrogeologic transport in a wet-rock repository, we adopt as a performance criterion the average yearly contribution of 10^{-4} Sv/yr to the lifetime dose commitment, corresponding to about 10 percent of the average yearly dose from natural background radiation.

The time-dependent source terms for calculating hydrogeological transport to the environment [30,31] are obtained from the initial inventories of Table 1 and the fractional dissolution rates of Table 2. Sorption of a dissolved radionuclide onto rock reduces its transport velocity below the groundwater velocity and provides more time for radioactive decay before the radionuclide reaches the biosphere. The ratio of the groundwater velocity to the radionuclide transport velocity is the retardation constant. Values [2] are given in Table 3.

Hydrologic data for different rock types [2] appear in Table 4. For given dissolution rates and transport distance, measured by the water travel time, the concentrations of released radionuclides depend also on the volumetric flow rate of underground or surface water which is transporting the contaminant plume from the repository. The volumetric flow rate of contaminated groundwater begins as the small amount flowing through the repository, and it increases as the contaminated water is diluted with interflows from aquifers in the surrounding media. No distance-dependent data on the flow rates of potentially contaminated groundwater were available for the generic or proposed repository sites considered in this study, so a constant value was adopted for each of the rock types.

The data for a site in natural salt are not expected flows, because natural salt contains no moving groundwater. They result from assuming that an aquifer in surrounding non-salt strata becomes diverted, dissolves and flows through the salt, dissolves wastes, and joins an aquifer that then flows eventually to the surface. The resulting salinity of the contaminated aquifer flow will affect the retardation constants of certain radionuclides, particularly cesium.

Also shown in Table 4 are volumetric flow rates of rivers that may be typical of flowing surface water into which the contaminated groundwater discharges. Dilution by the river flow further reduces the concentration of radionuclides in the biosphere.

Because of the slow dissolution rates of waste, the concentrations of released radionuclides vary little over a human lifetime, so these predicted concentrations can be multiplied by the dose conversion factors in Table 5 to obtain the average annual contribution to the lifetime dose commitment for a maximally exposed individual, a person whose entire intake of water and food is assumed to have been contaminated by radionuclides from the geologic repository. These dose factors were derived from the pathway and intake calculations of Napier, et al [40], but with modifications to the data for radium-226, lead-210, and neptunium-237 to conform with recent recommendations for these radionuclides [41,42].

Time-dependent radiation doses from groundwater transport

To illustrate the phenomena and time scales of geologic isolation, we assume a repository with the sorptive properties of basalt, a water travel time to the biosphere of 1,000 years, and congruent dissolution at a fractional rate of 10^{-4} /yr. Figure 2 shows the calculated relative contributions to the average annual dose to maximally exposed individuals. The actual radiation doses depend in magnitude on the volumetric flow rate of water carrying the released radionuclides and are presented later.

The first groundwater carrying dissolved carbon-14 and iodine-129 will take an average time of 1000 yr to carry these nonsorbing radionuclides to the 1 km location assumed in this calculation, but because of dispersion some of the contaminated water will arrive earlier, causing rounded edges of the fronts of the release bands. The release of carbon-14 and iodine-129 continues for a time given by the dissolution time, here assumed to be 10,000 years, the reciprocal of the fractional release rate. After 11,000 years, and neglecting some delayed transport from dispersion, all the nonsorbing radionuclides originally in the waste will have either decayed or will have discharged into the environment.

Because of its long half life of 17 million years, and for waste-dissolution times of the order of 10,000 years, all the iodine-129 originally in the waste discharges to the environment, and this will hold for water travel times orders of magnitude greater than that assumed in Figure 2. Even though some of the iodine may be ingested by humans for brief human-retention periods, it ultimately reaches the ocean, where it is further diluted and accumulates until it finally decays. Technetium-99, with a half life of 2.12×10^5 years, suffers much the same fate, assuming the same waste dissolution time and negligible sorption.

Dispersion causes a greater rounding and attenuation of the concentration bands of later arriving radionuclides, such as technetium-99 and neptunium-237.

Neptunium-237 does not appear appreciably in the environment until a time given by the product of the water transport time and its retardation constant of 100, i.e., 10^5 years. Because of its long half life of 2.14 million years and because of the low retardation constant, neptunium-237 is predicted to reach the 1-km environment in concentrations large enough to result in a relative radiation dose greater than from any other actinide. About 97 percent of the neptunium-237 in the waste would reach the environment from the hypothetical repository illustrated here, and most of this would ultimately reach the ocean.

Adopting retardation coefficients of 500 for plutonium and americium, these elements will not appear in the assumed environment until about 5×10^5 years, long enough for all their important radioisotopes to decay.

For the assumed 1000-year water travel time of Figure 2, 1602-yr radium-226 appears in the environment after 50,000 years, as a result of the transport of 2.47×10^5 -yr uranium-234, most of which is formed by the earlier decay of americium-242m, curium-242, plutonium-238 in the waste, according to the decay chain shown in Figure 3. The release band of radium-226 is broad, and it discharges to the environment occurs over a long time because of the simultaneous migration of its long-lived parents uranium-234 and thorium-230. For much longer water travel times radium-226 will discharge to the environment at a lower rate, a result of the transport of 4.51-billion-year uranium-238 and its daughters.

The discharge of radium-226 is accompanied by the discharge of its decay daughter lead-210, with a half life of 21 years. Although radium-226 and lead-210 will migrate through geologic media at activity concentrations in constant ratio given by the inverse ratio of their retardation coefficients [27], these two radionuclides can follow different food-chain pathways once they reach the environment. They are treated separately in the dose calculations.

Many other radionuclides that must be considered in analyzing the performance of a geologic repository are shown in the later graphs, but they are omitted from Figure 2 for simplicity.

Effects of water travel time and solubility-limited dissolution

Because the peak concentrations of released radionuclides occur over many human lifetimes, as illustrated in Figure 2, the

adequacy of long-term isolation can be measured by comparing the peak annual doses to the performance criterion of 10^{-4} Sv/yr. Dissolution is assumed to begin shortly after the repository is completely filled with waste from fuel reprocessing, and all waste packages are assumed to undergo dissolution at the same time and rate. The solubility-limited dissolution rates of Table 2 are applied individually to the radionuclides to calculate the peak individual doses. The dissolution rate of cesium is not solubility limited, so a fractional dissolution rate of 10^{-4} /yr is assumed. The effects of variations in the cesium dissolution rate are considered in the next section.

Instead of adopting the groundwater travel times quoted in Table 4 for various media and sites, we adopt the water travel time to the biosphere as a parameter in the calculations. Long-term radiation doses are estimated below for conceptual repositories in basalt, granite, salt, and tuff. The purpose is to show general features that affect long-term releases for each rock type. More detailed calculations would be required to fully evaluate the merits of the proposed sites that are included in this study.

Effect of dissolution rate and dispersion on cesium transport
 --- All the important radionuclides in reprocessing wastes other than cesium-137 can be limited in their dissolution rates because of the low solubilities of their compounds. We can only estimate the range of dissolution rates of cesium, since the solubilities of its usual compounds in water are too great to limit its dissolution rate.

A lower limit to the dissolution rate of cesium would be the dissolution rate of the waste matrix, estimated in Table 2 to be 1.1×10^{-6} /yr. However, some laboratory experiments show that cesium can diffuse preferentially through the waste solid. Cesium diffusion in solid waste, evident in some laboratory tests, could increase its dissolution rate, but there is no valid basis for predicting whether solid diffusion of cesium persists for the times of interest in a repository. Based on laboratory data for borosilicate glass, Crandall [43] quotes a cesium fractional release rate of 1.6×10^{-3} /yr. From the diffusion parameters suggested by the data of Godbee and Joy [44] we extrapolate to times of a few hundred years after the beginning of dissolution, and we estimate a steady-state fractional dissolution rate of cesium of 7×10^{-3} /yr for the waste-form dimensions given in Table 5. If the waste form were the ceramic Synroc, instead of borosilicate glass, laboratory data [45] show that the cesium fractional release rate may be as low as about 10^{-7} /yr.

In Figure 4 we show peak radiation doses from cesium-135 in a basalt repository that would result from the range of dissolution rates of cesium, from 1.1×10^{-6} /yr to 1.6×10^{-3} /yr. If dispersion is negligible, then the effect of uncertainties in cesium dissolution rate will be large and important, and cesium-

135 could be the main contributor to radiation dose for water travel times up to 10^5 yr. However, with an axial dispersion coefficient as large as $50 \text{ m}^2/\text{yr}$, cesium-135 is important only within the near-field of the repository. The three dispersion curves of Figure 4 coalesce at water travel times greater than about 10^3 yr. As illustrated here, the far-field peak concentration of a radionuclide migrating under the influence of dispersion can be independent of the dissolution rate of that radionuclide, if the migration time is much longer than the dissolution time. The greater near-field concentration resulting from a higher dissolution rate is more rapidly attenuated by dispersion because of the relative short time period over which the release occurs, and the resulting far-field concentration becomes identical to that from a lower dissolution rate.

Because of dispersion during hydrogeologic transport, the actual dissolution rate of cesium from the waste may be unimportant for repository sites wherein cesium retardation is as great as that adopted here for basalt. This insensitivity of cesium dose to dissolution rate is important for repositories in basalt, granite, or tuff, because of the high retardation coefficients and high dispersion during the water travel times of interest. It is not as useful for more weakly sorbing radionuclides, and it will not be as useful for cesium released from a salt repository, where sorption and dispersion of cesium are not as great as in basalt, granite, and tuff.

Performance of a repository in basalt -- The calculated peak individual doses for a repository in basalt are shown in Figure 5. Selenium-79, technetium-99, and tin-126 are no longer principal contributors to the dose. The solubility-limited dissolution rate of uranium reduces the radiation dose from uranium-234, but its most important effect is on the radiation dose from the decay daughters radium-226 and lead-210. Radium-226 and its precursor thorium-230 will not be present in enough mass for their solubilities to be limiting, but the low dissolution rate of the chain mother, uranium, means that the only way for appreciable concentrations and doses from radium-226 to appear in the environment is by transport of 80,000-yr thorium-230 directly from the waste. This is possible only for short water travel times, of the order of a few hundred years. Thus, when uranium is solubility limited, radium-226 is a principal contributor to the individual dose only for water-travel times of a few hundred years and less (cf. Figure 5).

Based on these predictions, and based on the estimates [31,32] of a water travel time to the environment of 1.5×10^5 yr for a repository in basalt at Hanford, Washington, the maximum individual dose in the river water will be about 10^{-11} Sv/yr, a factor of ten million below the radiation dose criterion of 10^{-4} Sv/yr adopted for this study.

For the average groundwater travel time to the Columbia River of 1.5×10^4 yr estimated by Dove, et al. [34,35], the

maximum dose to the individual will be about 3×10^{-9} Sv/yr, over four orders of magnitude below the individual-dose criterion.

Figure 5 represents a preliminary estimate of the possible performance of a basalt repository. It indicates an approach towards analyzing repository performance that takes into account the technical performance of the isolation system of waste package, geology, and hydrology for each of the important radionuclides. It suggests many technical features that will require verification through experiment and further analysis. Many features of the present analysis may conservatively predict greater doses than will occur. There are also many uncertainties and other phenomena, not taken into account in the present analysis, that must be considered before the margin of uncertainty in the predicted dose can be determined. Such analysis is beyond the scope of this study. However, the large margin between the calculated dose in river water and the goal of 10^{-4} Sv/yr may sufficiently allow for these uncertainties.

Performance of a repository in granite -- The solubility-limited dissolution rates of Table 2 are adopted here for the granite repository, and the predicted doses are shown in Figure 6. Because the controlling radionuclides have the same estimated retardation constants for granite as for basalt, the calculated doses for granite behave in the same way as for basalt, but the magnitude of doses in the surface water for the granite site are about 30-fold higher, because of the lower river flowrate assumed for the granite site. If the water travel time to the environment for the granite site is about 1000 years, the maximum predicted radiation dose is from carbon-14. If the water travel time is 10^5 yr, a maximum dose rate of about 3×10^{-9} Sv/yr occurs in the surface water for the granite site, a factor of 3×10^4 below the radiation dose criterion of 10^{-4} Sv/yr.

Performance of a repository in salt -- The solubility-limited dissolution rates of Table 2 are adopted for a salt repository, although it might be expected that the geochemical environment could result in somewhat greater solubilities and dissolution rates if groundwater were to intrude into salt. The estimated radiation doses for a salt repository are shown in Figure 7. Here cesium-135 dominates the dose for water travel times greater than 10^5 yr. Determining the appropriate dissolution rate of cesium-135 is specially important for a salt repository. For the cesium retardation coefficient adopted here, cesium dispersion is not enough to result in the insensitivity of predicted doses to the dissolution rate of cesium.

It is inappropriate to compare the calculated doses for a salt repository with the individual dose criterion of 10^{-4} Sv/yr. The latter is adopted for expected events. Groundwater is not expected to flow through salt, and hydrogeologic transport of radionuclides from the salt repository is not probable. It is normal practice to adopt higher values of radiation dose criteria

for improbable events, as would result from a probability-weighted risk analysis.

Performance of a repository in tuff -- Adopting the solubility-limited dissolution rates of Table 2, the calculated doses for a tuff repository are shown in Figure 8. The relative values of the doses from individual radionuclides vary with water travel time in the same way as for the basalt and granite repositories. No estimates of groundwater flowrate for a tuff site were available for this study, and it appears that there is no surface water into which potentially contaminated groundwater can discharge. Attention must be focused on possible use of the potentially contaminated groundwater by humans, for potable water and irrigation.

If 4300 yr is the time for groundwater to travel from a repository in saturated tuff to an off-site well, the radiation dose will be dominated by carbon-14 or, depending on its dispersion coefficient, by cesium-135. If 2.1×10^4 yr is the time for groundwater to travel to the same off-site location from a repository in unsaturated tuff, the carbon-14 dose will be attenuated so that it is comparable with the predicted dose from neptunium-237. If the groundwater flow rate is within the range of flow rates considered in this study, the individual dose from carbon-14 in groundwater would be between one and two orders of magnitude greater than the performance criterion adopted for this study. The probability of future use of this contaminated groundwater must be taken into account. If the generally arid area of the tuff site is expected to persist into the distance future, it seems that the potentially contaminated groundwater is likely to be used.

This performance analysis of a tuff repository illustrates differences between an individual dose criterion and a radioactivity release limit, the latter appearing in a standard recently proposed by the Environmental Protection Agency [4]. According to the present calculations a repository in Nevada tuff would meet EPA's proposed limits on the amount of activity that could be released during the first 10,000 years. Yet, with the parameters adopted for the calculations herein, a tuff repository may not meet the individual dose criterion. EPA's proposed standard, and numerical criteria recently adopted by the Nuclear Regulatory Commission [47] to implement the EPA standard, are not concerned with the effects of water flow rates, nor are they concerned with concentrations and individual doses from radionuclides in groundwater and surface water. Deciding what criterion to use in assessing the performance of geologic repositories will aid present and future programmatic decisions.

Radiation doses for a repository loaded with unprocessed spent fuel

The calculated radiation doses for a basalt repository completely loaded with unprocessed spent fuel are shown in

Figure 9. In estimating the solubility-limited dissolution rates for spent fuel we assume that the solubilities for the stable species in groundwater are those listed in Table 2. For simplicity, we neglect the larger waste surface area per unit mass of contained fission products for a spent-fuel waste package as compared with the waste package containing reprocessed high-level waste, so the resulting mass dissolution rate of the parent radionuclide is the same as for reprocessed waste. The predicted doses are shown in Figure 9. The calculated doses from neptunium-237 are the same for spent fuel as for reprocessed waste (cf. Figure 5). This is also true for selenium-79, americium-241, and the other solubility-limited species within the high-level waste package.

Because uranium is solubility limited, radium-226 migrates appreciably from the repository only as a result of the migration of thorium-230, which is not at a high enough concentration to be solubility limited. Thus, the greater amount of uranium-234 in spent fuel creates more thorium-230 and much greater concentrations of radium-226 in groundwater, for water travel times less than about a thousand years.

If the dissolution rate of cesium is the same for spent fuel as for reprocessed waste, the dose rate from cesium-135 will be the same. Laboratory data suggest a greater dissolution rate for cesium in spent fuel, but cesium dispersion, as illustrated in Figure 4, should make the cesium dose rate for spent fuel insensitive to dissolution rate for water travel times greater than a few hundred years.

Because spent fuel is not reprocessed, each high-level waste package contains its proportion of the carbon-14 and iodine-129 in the repository. The ratio of surface area to the inventory of each of these two radionuclides in each spent-fuel package results in no solubility limits on their dissolution. Their dissolution rates are likely to be determined by the kinetics of dissolution and restructuring of the uranium-dioxide fuel. Assuming a fractional release rate of 10^{-4} /yr for carbon-14 and iodine-129, we predict that carbon-14 will be a large contributor to the dose rates for groundwater travel times less than about 10^5 yr, and iodine-129 will be the main contributor to radiation doses at longer times. Disposing of spent fuel as waste not only precludes the option of designing solubility-limited waste packages for carbon-14 and iodine-129, but it also precludes the options of nongeologic disposal of these radionuclides.

The differences between spent fuel and reprocessed waste, as illustrated here for a basalt repository, will apply as well to the other repository media.

Effect of repository heating

Even if a corrosion-resistant overpack is included in the

waste package to protect the waste forms from exposure to groundwater during the thermal period, some waste forms will still be exposed because of statistically expected failures of some waste packages. A greater number of waste forms will be exposed to groundwater during the thermal period if the overpack is not used. Although the U. S. Nuclear Regulatory Commission [46,47,48] concluded that temperature effects are important and should be avoided by an overpack, no calculations or experiments were available to justify those conclusions. In the absence of data or analyses from other sources, we have made our own estimates of the effect of higher-temperature exposure on dissolution rates and repository performance.

When the repository temperatures are higher than ambient, solubility-limited dissolution rates can increase because of the increased solubilities and liquid diffusion coefficient, and because the increased local velocities of groundwater from thermal-convection flow can decrease the boundary layer thickness. The effects of temperature on the solubility of silica and on the diffusion coefficient in the liquid are shown in Table 6.

Temperatures affecting solubilities during the thermal period are the time-dependent rock temperatures at the emplacement-hole surface, shown in Figure 10. The basalt project has supplied analyses of the thermal-convective flows through a repository [33], including time-dependent temperatures [49], potential gradients, pore velocities, and volumetric flow rates through the repository rock. During the thermal period the thermally induced potentiometric gradient far outweighs the ambient gradient, and the local pore velocities in the host rock increase from the ambient value of 3.2×10^{-3} /yr to as high as 0.64 m/yr. The time for water to travel to the edge of the host rock decreases from 6600 yr to 33 yr. These host-rock travel times are small compared with the total travel time to the river, estimated to be 1.9×10^5 yr without repository heating and 2.0×10^5 yr with heating. Heating is calculated [33] to increase the water travel time because buoyancy forces the exterior flow into strata of lower permeability.

Even during the period of repository heating the groundwater velocities are low enough that we must analyze diffusion-controlled transport through the concentration boundary layer. However, the steady-state dissolution rate of Equation (2) cannot be used, because of the time-dependent boundary condition and diffusion coefficient and because steady-state has not been reached. Chambre [50] recently solved this time-dependent dissolution rate for a sphere. Because of the transients, the dissolution rate depends on the local retardation coefficient, as shown in Figure 10. These calculations were made using the temperature-dependent solubility of amorphous silica, but the normalized results will apply to other constituents if their heats of solution are the same as that of silica. A local temperature as high as 250°C , together with transient effects,

can increase the dissolution rate about 150-fold above the long-term ambient rate.

The dissolution rates of Figure 10 can be used to calculate the far-field concentration $N(z,t)$ from one-dimensional advective transport, using the general solution presented elsewhere [30]. The results for neptunium-237 at 100 meters from the repository, with an assumed retardation coefficient of 100, are shown in Figure 11 for various values of the hydrodynamic dispersion coefficient. If there were no dispersion, the greater dissolution rate at 1 year would result later in a concentration at 100 meters 150-fold greater than the concentration from steady-state dissolution at ambient temperature. This concentration spike rapidly disappears, because later arrivals at 100 meters reflect lower-temperature dissolution and less-steep concentration gradients at the waste surface.

Because the thermal spike in the dissolution rate lasts only a few hundred years, dispersion during groundwater transport broadens and attenuates this spike. Increasing dispersion causes earlier arrival of the contaminant, destroys the far-field concentration spike, and results in far-field concentrations only slightly above those predicted for steady-state dissolution at ambient temperatures. This suggests that exposing high-level waste to groundwater during the thermal period may result in only small increases in the far-field concentrations above those predicted for the later steady-state dissolution at ambient temperature.

These results question the need for the corrosion-resistant canister that has been proposed to delay dissolution until repository heating has subsided.

In conclusion

Based on these predictions, several geologic media are expected to result in radiation doses to the maximally exposed individual that are far below the 10^{-4} Sv/yr criterion adopted for this study. These long-term individual doses from hydrogeologic transport of dissolved radionuclides to the biosphere are expected for wet-rock repositories, but are not expected for repositories in natural salt.

Developing and validating techniques for predicting long-term performance of geologic repositories is essential to the successful completion of the national program for geologic isolation. Although the actual amount of radionuclides that can reach the biosphere is small, environmental releases are predicted to occur for hundreds of thousands of years. Long-term performance of repositories cannot be demonstrated by real-time laboratory experiments and pilot testing. Extrapolating laboratory and field data and predicting long-term releases to the environment must rely on theories that are sound and verified.

The chemical engineering principles of mass transfer by diffusion and convection have recently been applied to predict the rate of dissolution of low-solubility radioactive constituents in solid waste. The theory predicts that most of the important constituents in high-level and transuranic waste from fuel reprocessing are of low enough solubility that their dissolution rates will be low and will be controlled by the rate of mass transfer of the dissolved constituents through the concentration boundary layer exterior to the waste surface. The theory shows that wastes of separated carbon-14 and iodine-129, if emplaced in a geologic repository, can be designed so that their rates of solubility-limited dissolution are suitably low. Higher dissolution rates of some of the important radionuclides are predicted if unprocessed spent fuel is emplaced as solid waste in repositories, and greater long-term releases to the biosphere are predicted.

The theory of mass-transfer-controlled dissolution predicts that the dissolution of solid waste during repository heating may be several orders of magnitude greater than the longer-term dissolution rate at ambient temperature. However, because of hydrodynamic dispersion during transport through the rock media, this transient dissolution rate is predicted to result in no appreciable increase in the dose rate to the maximally exposed individual when the radionuclides reach the biosphere.

The validity of these mass-transfer theories should be tested in laboratory experiments simulating waste canisters surrounded by saturated porous and fractured media.

The chemical engineering principles of chromatographic transport have been extended to the simultaneous advective transport of radionuclides in decay chains. Some important radionuclides, e.g., radium-226 and lead-210, reach the environment by the advective transport of their precursors, so accurate calculation of decay-chain transport is important.

Theoretical predictions of long-term repository performance should be extended to include details of site-specific hydrology and geochemistry. They must include analyses of the uncertainties in data and predictive techniques, and they must consider other modes by which radionuclides can reach the biosphere, in addition to hydrogeologic transport that is the subject of this study.

The choice of different rock media and sites, as well as the choice of technology for designing repositories and waste packages, depend on the performance criterion that is adopted.

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Table 1 Radionuclide Inventories in Wastes
from Fuel Reprocessing a/

Radionuclide	Half life, Yr	Inventory Bq
C-14	5.73×10^3	5.73×10^{15}
Se-79	$\leq 6.5 \times 10^4$	1.46×10^{15}
Sr-90	2.77×10^1	2.68×10^{20}
Zr-93	1.5×10^6	6.68×10^{15}
Tc-99	2.12×10^5	4.84×10^{16}
Sn-126	1×10^5	2.88×10^{15}
I-129	1.7×10^7	1.16×10^{14}
Cs-135	3×10^6	1.28×10^{15}
U-234	2.47×10^5	4.16×10^{13}
U-238	4.51×10^9	1.16×10^{13}
Np-237	2.14×10^6	1.16×10^{15}
Pu-238	8.6×10^1	4.26×10^{16}
Pu-239	2.44×10^4	1.16×10^{16}
Pu-240	6.58×10^3	1.95×10^{16}
Pu-241	1.32×10^1	4.57×10^{18}
Pu-242	3.79×10^5	6.50×10^{13}
Am-241	4.58×10^2	7.03×10^{17}
Am-243	7.95×10^3	6.31×10^{16}
Cm-242	4.46×10^{-1}	7.29×10^{19}
Cm-244	1.76×10^1	5.55×10^{18}
Cm-245	9.3×10^3	7.68×10^{13}
Cm-246	5.5×10^3	1.06×10^{14}

a/ Total repository waste from 10^5 Mg uranium in light-water reactor fuel. Assumes 0.5% of U + Pu become high-level waste and 0.5% of U + Pu become transuranic waste. Calculated for 165 days after reactor discharge.

Table 2. Calculated fractional release rates for borosilicate glass waste at 20°C

Waste cylinder: radius = 0.152 m, length = 2.46 m, fission-product and actinide oxides from 460 kg of uranium in fuel, groundwater pore velocity = 1 m/yr.

Constituent	Waste concentration, g/cm ³	Solubility ^a , g/cm ³	Fractional release rate	
			Calculated ^b yr ⁻¹	Observed ^c yr ⁻¹
SiO ₂	1.6	5×10^{-5}	1.1×10^{-6}	1.6×10^{-3}
Tc	1.92×10^{-3}	1×10^{-9}	2×10^{-8}	
U	1.22×10^{-2}	1×10^{-9}	4×10^{-9}	1.5×10^{-6}
Np	1.92×10^{-3}	1×10^{-9}	2×10^{-8}	6.6×10^{-4}
Pu	1.15×10^{-4}	1×10^{-9}	4×10^{-7}	2.6×10^{-5}
Am	3.56×10^{-4}	1×10^{-10}	1×10^{-8}	2.7×10^{-5}
Se	1.40×10^{-4}	1×10^{-9}	3×10^{-7}	
Sn	9.40×10^{-5}	1×10^{-9}	5×10^{-7}	

a/ For amorphous SiO₂ [21]. Other solubilities are from Table 4.

b/ Calculated from Equation (1).

c/ Data of McVay, et al. [22] for IAEA-type leach tests, with periodic replacement of leachant.

Table 3. Solubilities and Retardation Factors ^{a/}

Element	Most Probable	Solubility, log ppm				Retardation Factor ^{b/}				
		Reducing: Eh=-0.2		Oxidizing: Eh=+0.2		Granite	Basalt	Tuff	Clay, Soil, Shale	Salt ^{c/}
		pH = 9	pH = 6	pH = 9	pH = 6					
Se	-3(?)	----	----	----	----	5 <i>50</i> 200	5 <i>50</i> 200	5 <i>50</i> 200	5 <i>50</i> 200	20 <i>200</i> 1,000
Sr	High	-0.2	High	-0.2	High	10 <i>200</i> 2,000	50 <i>200</i> 2,000	20 <i>200</i> 10,000	50 <i>200</i> 5,000	1 <i>10</i> 100
Zr	-4	-4	-6	-4	-6	500 <i>5,000</i> 30,000	500 <i>5,000</i> 10,000	500 <i>5,000</i> 10,000	500 <i>5,000</i> 50,000	300 <i>1,000</i> 5,000
Tc	-3	-10	High	High	High	1 <i>5</i> 40	1 <i>5</i> 100	1 <i>5</i> 100	1 <i>5</i> 20	1 <i>5</i> 20
Sn	.3(?)	-4	-4	-4	-4	100 <i>1,000</i> 5,000	100 <i>1,000</i> 5,000	200 <i>1,000</i> 5,000	200 <i>1,000</i> 5,000	10 <i>100</i> 1,000
Sb	-3(?)	----	----	----	----	10 <i>100</i> 1,000	10 <i>100</i> 1,000	10 <i>100</i> 1,000	10 <i>100</i> 1,000	5 <i>50</i> 500
I	High	High	High	High	High	1 <i>1</i> 1	1 <i>1</i> 50	1 <i>1</i> 1	1 <i>1</i> 1	1 <i>1</i> 1
Cs	High	High	High	High	High	100 <i>1,000</i> 10,000	100 <i>1,000</i> 10,000	60 <i>500</i> 10,000	200 <i>1,000</i> 20,000	1 <i>10</i> 2,000
Pb	-1	-1	0	-1	0	10 <i>50</i> 200	20 <i>50</i> 500	20 <i>50</i> 500	20 <i>50</i> 500	5 <i>20</i> 100
Ra	-2	-3	-1	3	-1	50 <i>500</i> 5,000	50 <i>500</i> 5,000	50 <i>500</i> 5,000	50 <i>500</i> 5,000	5 <i>50</i> 500
Th	-3	-4	-4	-4	-4	500 <i>5,000</i> 10,000	500 <i>5,000</i> 10,000	500 <i>5,000</i> 10,000	500 <i>5,000</i> 50,000	300 <i>1,000</i> 5,000
U	-3	-3	-5	High	High	10 <i>50</i> 500	20 <i>50</i> 1,000	5 <i>40</i> 200	50 <i>200</i> 5,000	10 <i>20</i> 60
Np	-3	-4	-4	-2	-1	10 <i>100</i> 500	10 <i>100</i> 500	10 <i>100</i> 500	10 <i>100</i> 400	10 <i>50</i> 300
Pu	-3	-5	-4	-5	-3	10 <i>200</i> 5,000	100 <i>500</i> 5,000	50 <i>200</i> 5,000	500 <i>1,000</i> 20,000	10 <i>200</i> 10,000
Am	-4	-8	-5	-8	-5	500 <i>3,000</i> 50,000	60 <i>500</i> 50,000	300 <i>1,000</i> 50,000	200 <i>800</i> 50,000	300 <i>1,000</i> 5,000
Cm	-3(?)	----	----	----	----	200 <i>2,000</i> 10,000	100 <i>500</i> 10,000	100 <i>500</i> 10,000	200 <i>2,000</i> 20,000	200 <i>1,000</i> 3,000

a/ From Krauskopf [2,23]

b/ Assumes pore volume/solid volume in surrounding media = 0.10.
Recommended values are in italics.

c/ For saline ground water in media surrounding salt.

Table 4. Hydrologic Parameters for Reference Repository Sites

Location	Repository Rock		Bedded ^b Salt	Domal ^c Salt	Granite ^d Generic	Tuff ^e	
	Basalt ^a Hanford, Wash.					Saturated Nevada Test Site	Unsaturated
Flow rate through repository (m ³ /yr)	17		40 to 400				
Flow rate of contaminated aquifer (m ³ /yr)	3.2x10 ⁴ (at 10 ⁴ yr)		1.3x10 ³	5.2x10 ⁵			
Flow rate of surface water (m ³ /yr)	1.1x10 ¹¹		3.2x10 ⁸	2.8x10 ⁹	3.8x10 ⁹		
Path lengths, repository to biosphere (km)	Rockwell estimates	PNL ^f estimates				6 (to well J13) 25 (to Lathrop Wells)	7 (to well J13) 26 (to Lathrop Wells)
Minimum	60	12	10 ³		1		
Maximum	80	16	10 ³		10 ²		
Mean	70	14	10 ²		10		
Water travel time to biosphere (yr)						1.2x10 ³ (to well J13)	2.2x10 ⁴ (to well J13)
Minimum	1.3x10 ⁴	1.3x10 ⁴	1x10 ⁴		10 ³	4.3x10 ³ (to Lathrop Wells)	2.5x10 ⁴ (to Lathrop Wells)
Maximum	1x10 ⁶	1.7x10 ⁴	1x10 ⁶		>10 ⁶		
Mean	1.9x10 ⁵	1.5x10 ⁴	1x10 ⁵	4x10 ⁴	10 ⁶		
Average pore velocity							
Emplacement rock (m/yr)	0.0032						0.03
Surrounding media (m/yr)	0.5	0.9	1	2		5.7	5.7
Effective porosity in aquifer	10 ⁻³ to 5x10 ⁻²	1.8x10 ⁻¹	0.10	10 ⁻⁵ to 3x10 ⁻²		0.18 (to Lathrop Wells)	

^aD. J. Brown and R. A. Deju [32,33]^bS. Goldsmith [36], except flow rate of contaminated aquifer is from G. E. Raines [38]. Data assume a salt repository breached by flowing water.^cData assume a salt repository breached by flowing water [2].^dData from [1], except flow rate of contaminated aquifer is from Cloninger and Cole [6].^eTyler [38].^fDove [34], Dove et al. [35].

Table 5. Average Lifetime Dose Rate per Unit Concentration of Radionuclides in Water

Radionuclide	Half-Life(yr)	Average Dose Rate per Unit Concentration ^a (Sv-m ³ /Bq-yr)	Ratio of Drinking Water Dose to Total Dose ^a
¹⁴ C	5.73×10^3	9.21×10^{-7}	1.22×10^{-4}
⁷⁹ Se	6.5×10^4	1.60×10^{-7}	4.27×10^{-4}
⁹³ Zr	1.5×10^6	4.83×10^{-13}	3.0×10^{-1}
⁹⁹ Tc	2.12×10^5	7.03×10^{-10}	1.0×10^{-2}
¹²⁶ Sn	1×10^5	2.77×10^{-8}	1.2×10^{-2}
¹²⁹ I	1.7×10^7	2.04×10^{-8}	7.07×10^{-2}
¹³⁵ Cs	3×10^6	5.26×10^{-8}	2.67×10^{-2}
²¹⁰ Pb	2.1×10^1	7.68×10^{-6} ^b	5.08×10^{-2}
²²⁵ Ra	4.05×10^{-2}	1.40×10^{-6}	1.16×10^{-1}
²²⁶ Ra	1.60×10^3	2.40×10^{-6} ^c	1.07×10^{-1}
²²⁹ Th	7.34×10^3	5.56×10^{-7}	6.84×10^{-2}
²³⁰ Th	8×10^4	8.03×10^{-8}	6.91×10^{-2}
²³³ U	1.62×10^5	3.80×10^{-8}	1.92×10^{-1}
²³⁴ U	2.47×10^5	3.80×10^{-8}	1.88×10^{-1}
²³⁸ U	4.51×10^9	2.91×10^{-8}	2.15×10^{-1}
²³⁷ Np	2.14×10^6	1.29×10^{-5} ^d	8.41×10^{-2}
²³⁹ Pu	2.44×10^4	9.80×10^{-9}	1.94×10^{-1}
²⁴⁰ Pu	6.58×10^3	9.80×10^{-9}	1.94×10^{-1}
²⁴² Pu	3.79×10^5	9.50×10^{-9}	1.85×10^{-1}
²⁴¹ Am	4.58×10^2	1.26×10^{-7}	4.3×10^{-2}
²⁴³ Am	7.95×10^3	1.23×10^{-7}	4.29×10^{-2}

a/ Derived from data Napier et al.[3] unless otherwise noted.

b/ Increased by a factor of 4.4 to allow for ICRP-30 corrections [5].

c/ Reduced by a factor of 90 to allow for ICRP-30 corrections [5].

d/ Increased by a factor of 200 to allow for ICRP-30 corrections [5].

Table 6. Effect of temperature on the solubility and liquid diffusion coefficient for silica

Temperature, °C	Solubility ^{a/} g/cm ³	Diffusion coefficient ^{b/} cm ² /sec
20	5.0×10^{-5}	1×10^{-5}
50	8.8×10^{-5}	2.0×10^{-5}
100	1.7×10^{-4}	4.5×10^{-5}
150	2.6×10^{-4}	7.9×10^{-5}
200	4.2×10^{-4}	1.2×10^{-4}
250	5.8×10^{-4}	1.6×10^{-4}

a/ Data of Fournier and Power [21].

b/ Estimated from the Stokes-Einstein equation: $D\mu T = \text{constant}$,
D = diffusion coefficient, μ = viscosity, T = absolute temperature

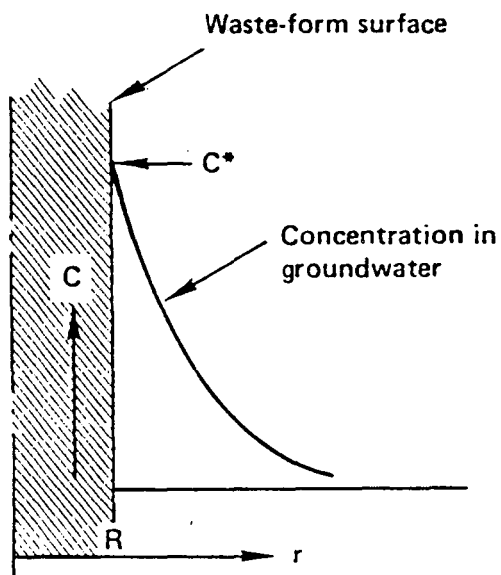
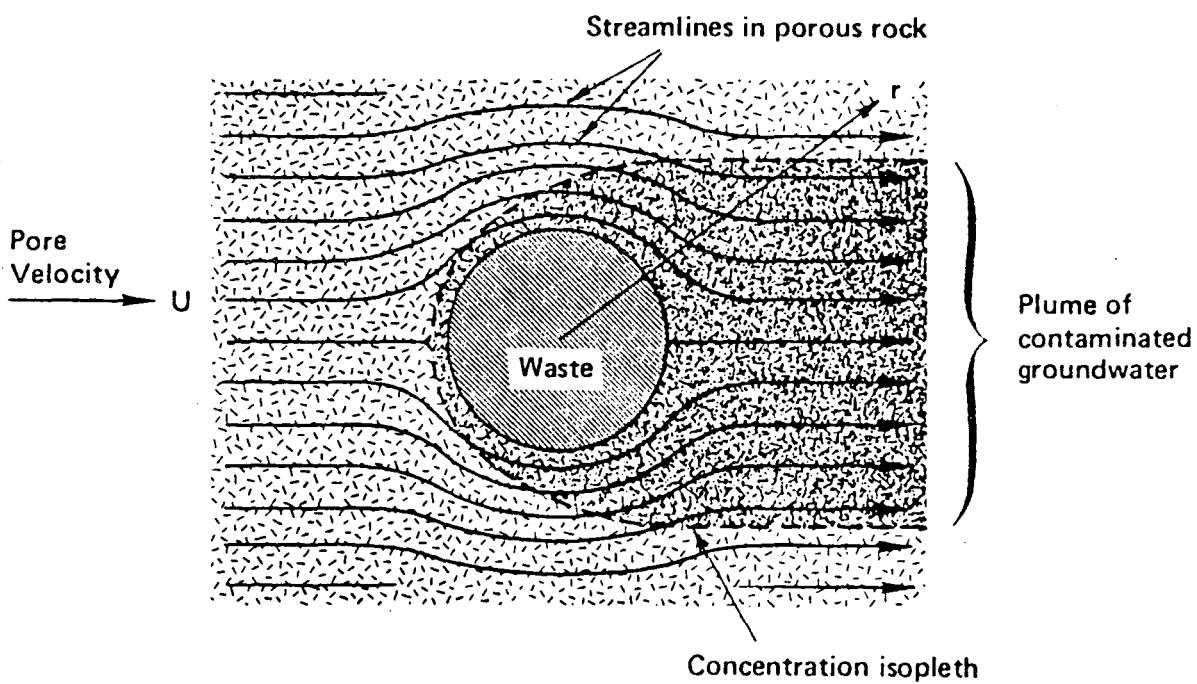


Figure 1. Velocity and concentration profiles for groundwater flowing across a waste cylinder.

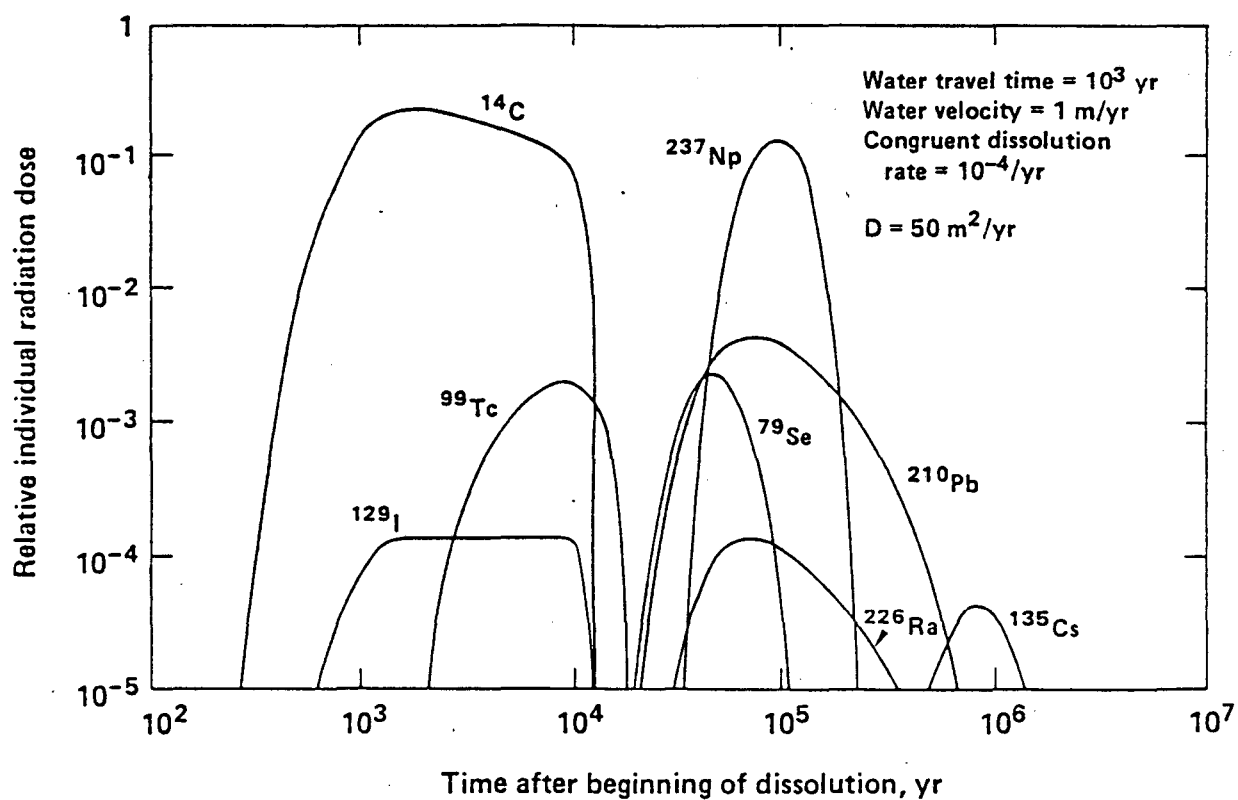


Figure 2. Relative individual radiation dose as a function of time for basalt, reprocessing waste, congruent dissolution

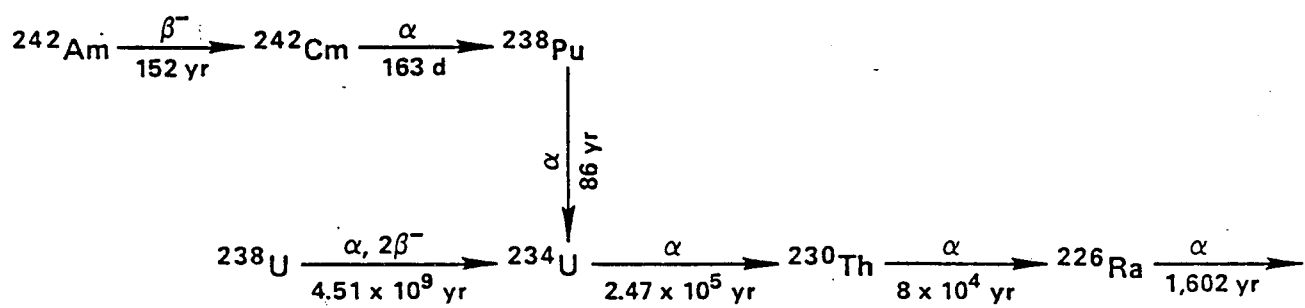


Figure 3. Decay chains leading to radium-226

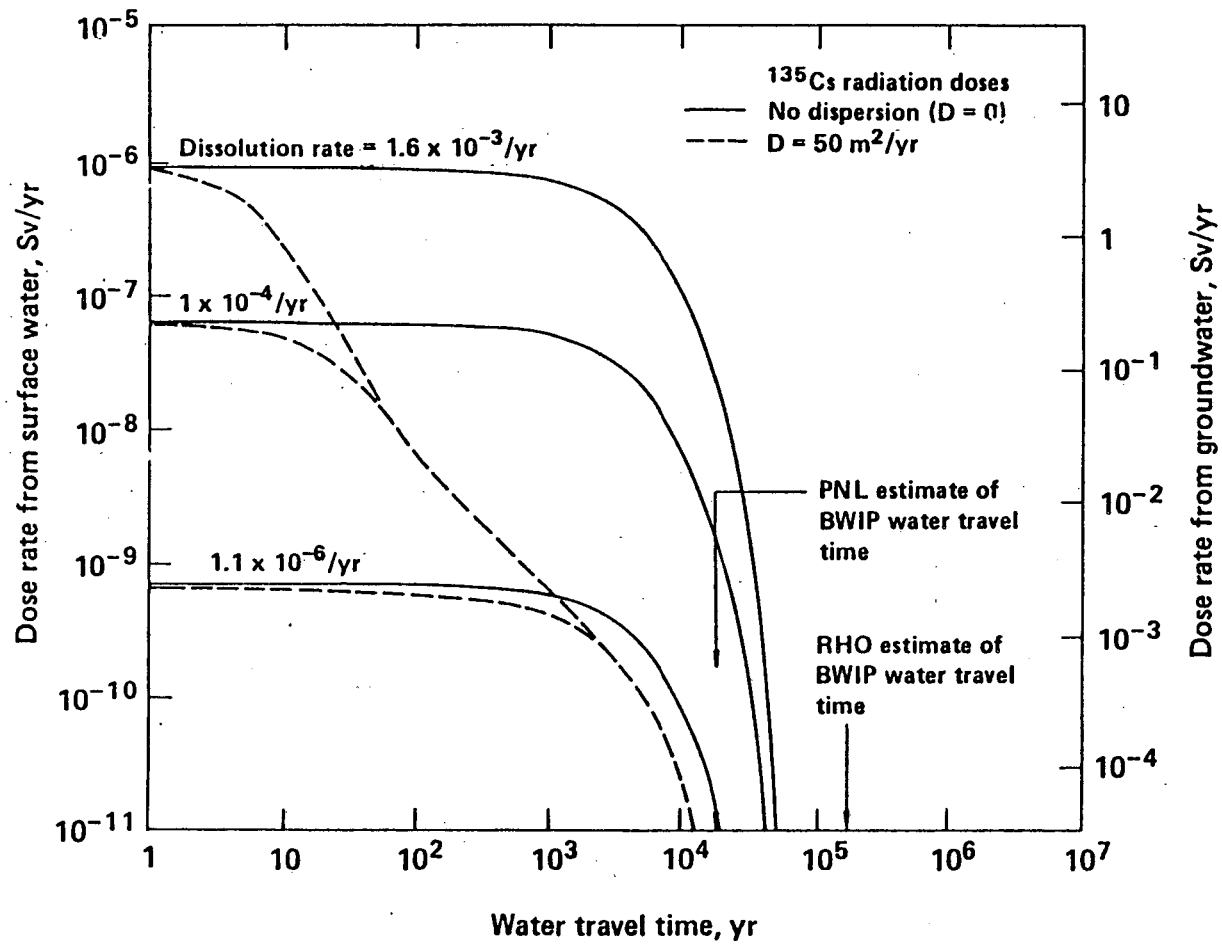


Figure 4. Individual dose from cesium-135 as a function of water travel time for basalt, effect of dissolution rate and dispersion, for reprocessing waste.

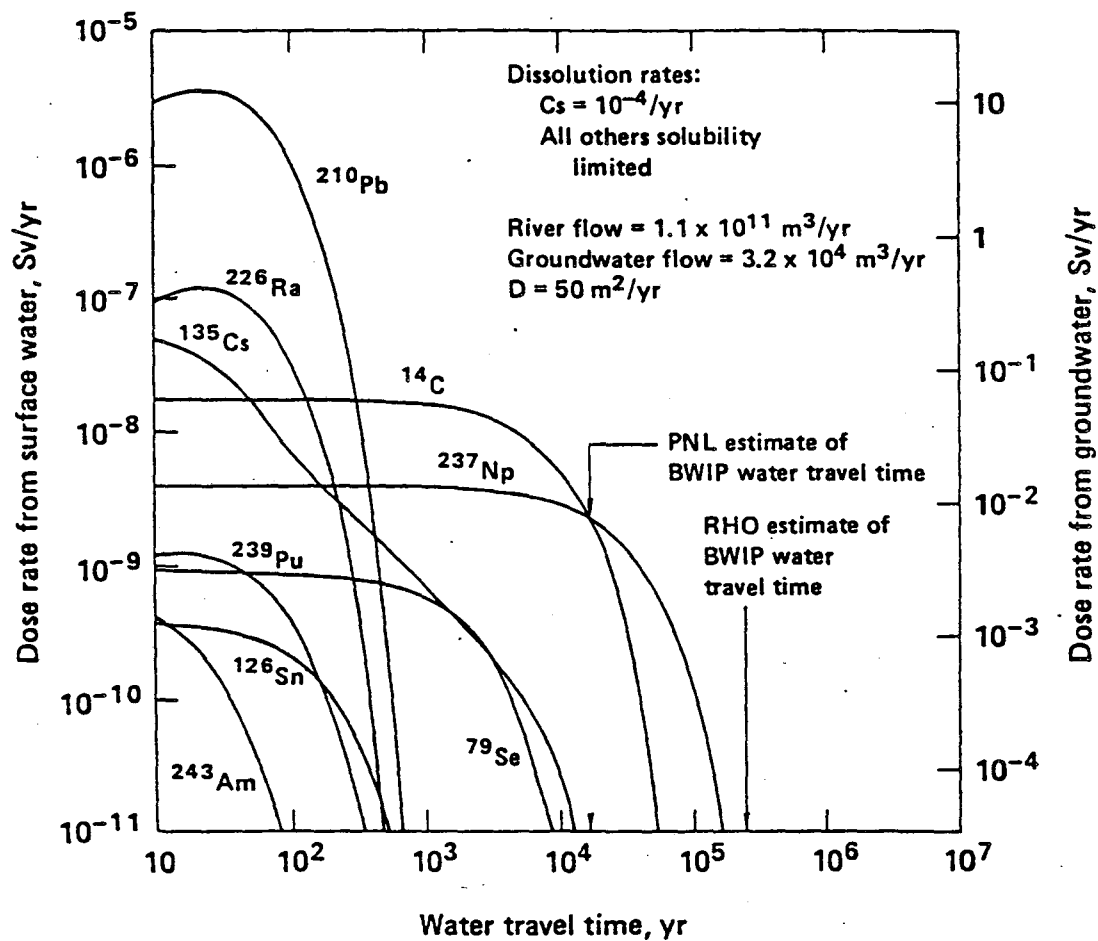


Figure 5. Individual radiation dose as a function of water travel time for basalt, reprocessing waste, solubility-limited dissolution.

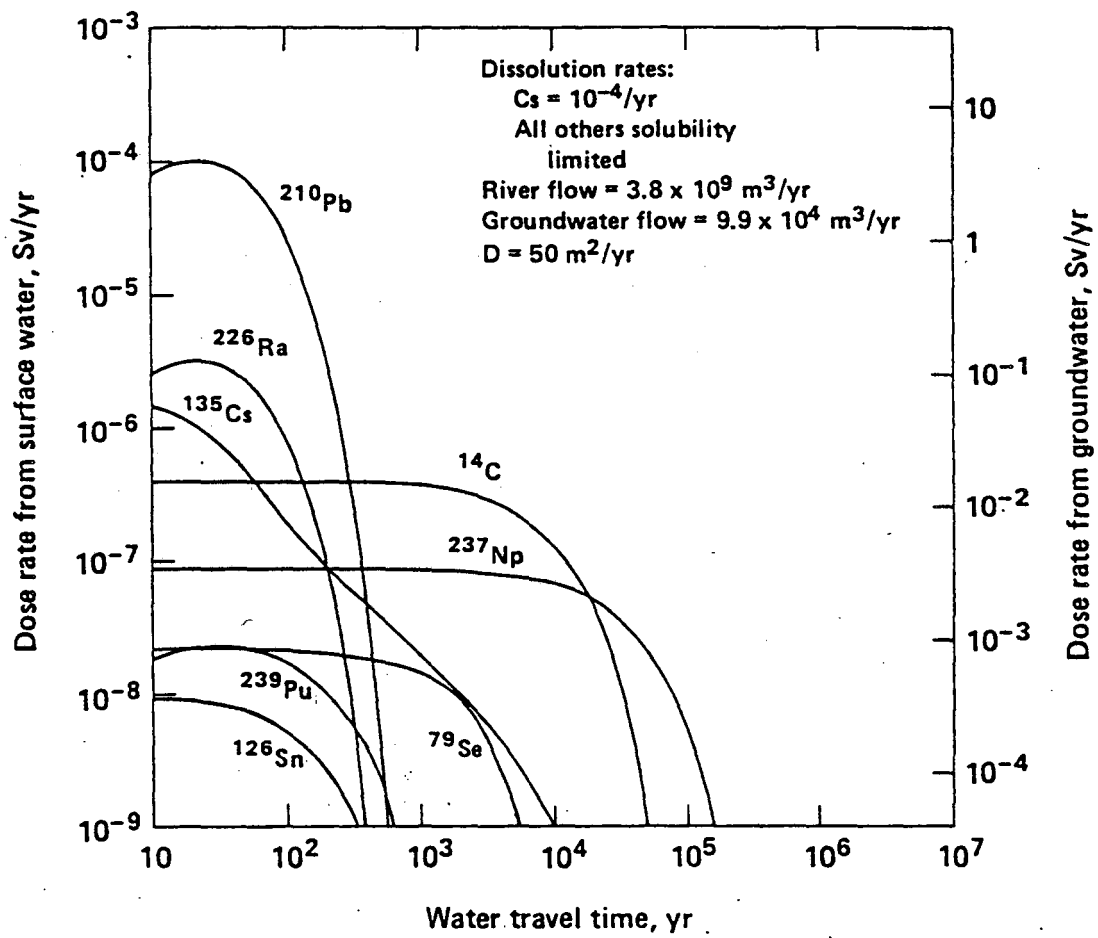


Figure 6. Individual radiation dose as a function of water travel time for granite, reprocessing waste, solubility-limited dissolution.

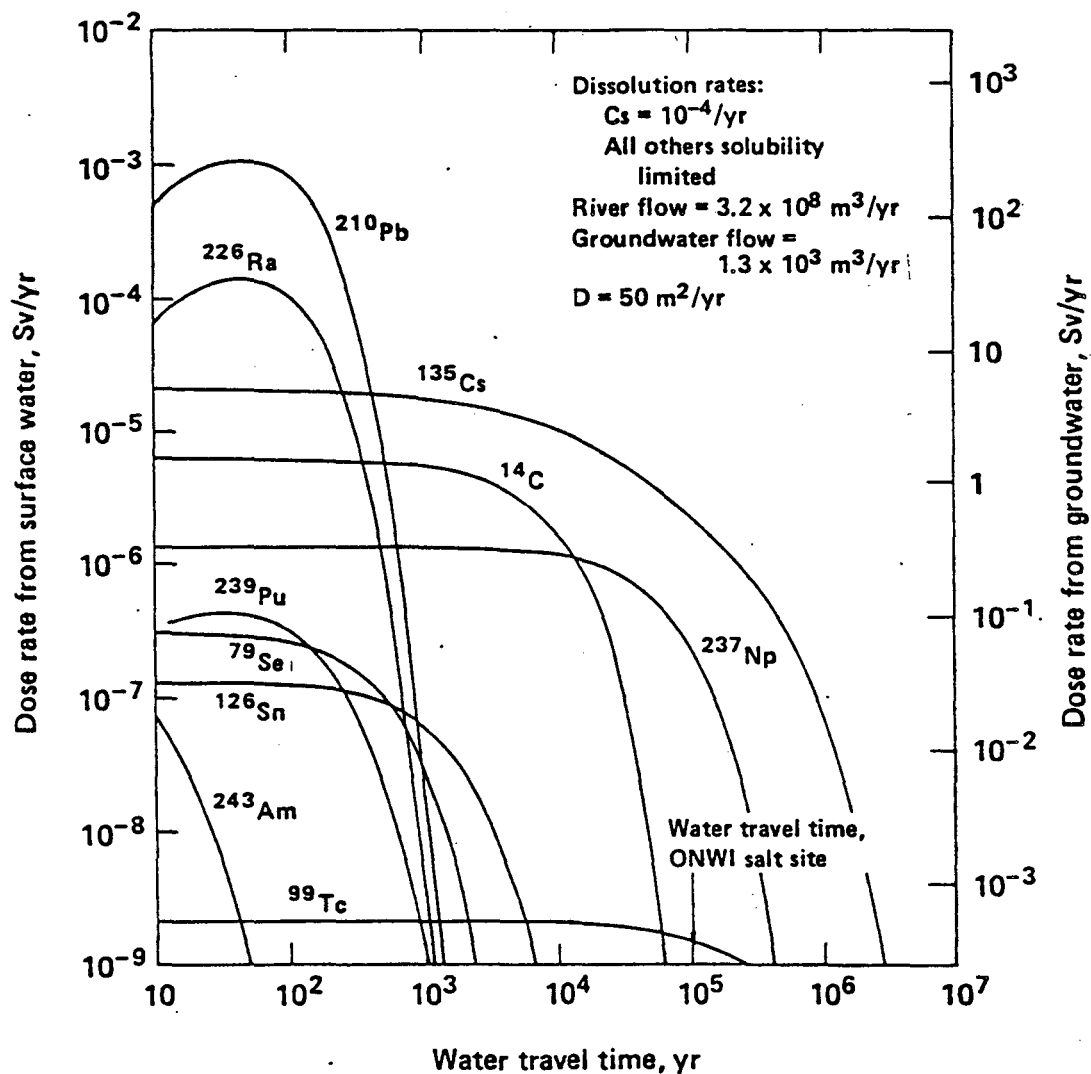


Figure 7. Individual radiation dose as a function of water travel time for salt, reprocessing waste, solubility-limited dissolution.

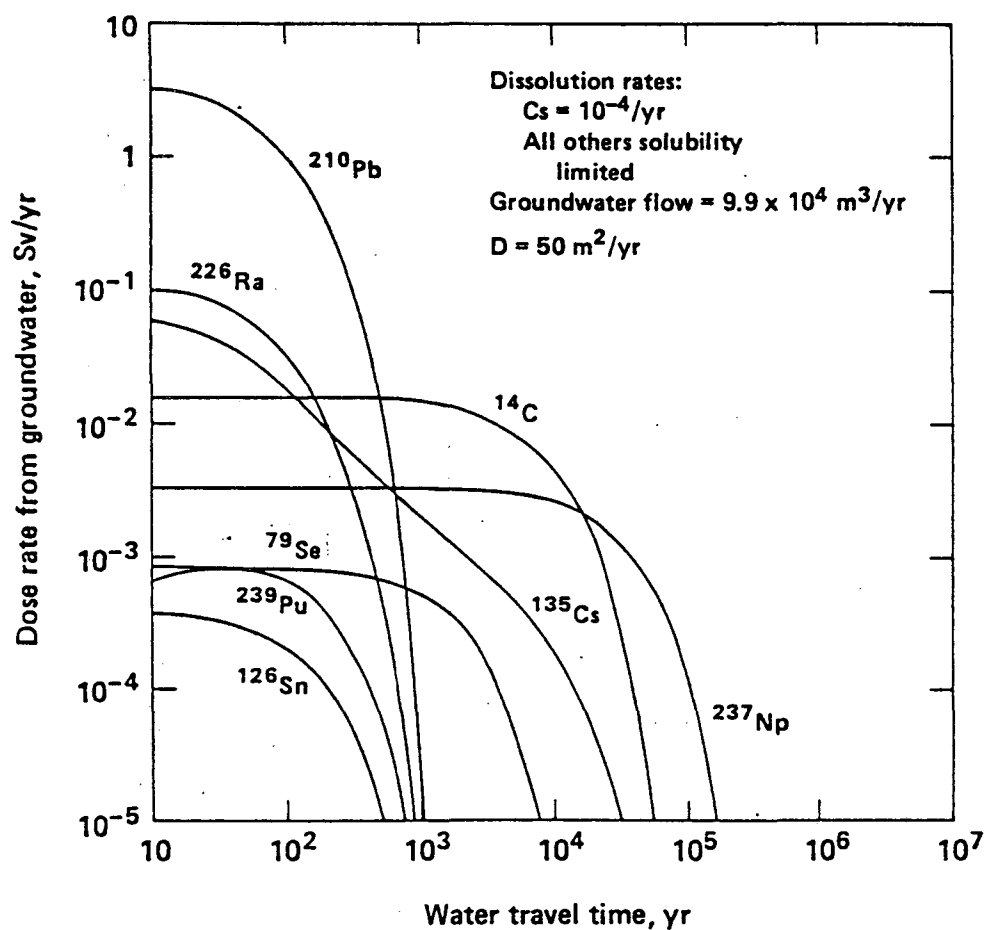


Figure 8. Individual radiation dose as a function of water travel time for tuff, reprocessing waste, solubility-limited dissolution.

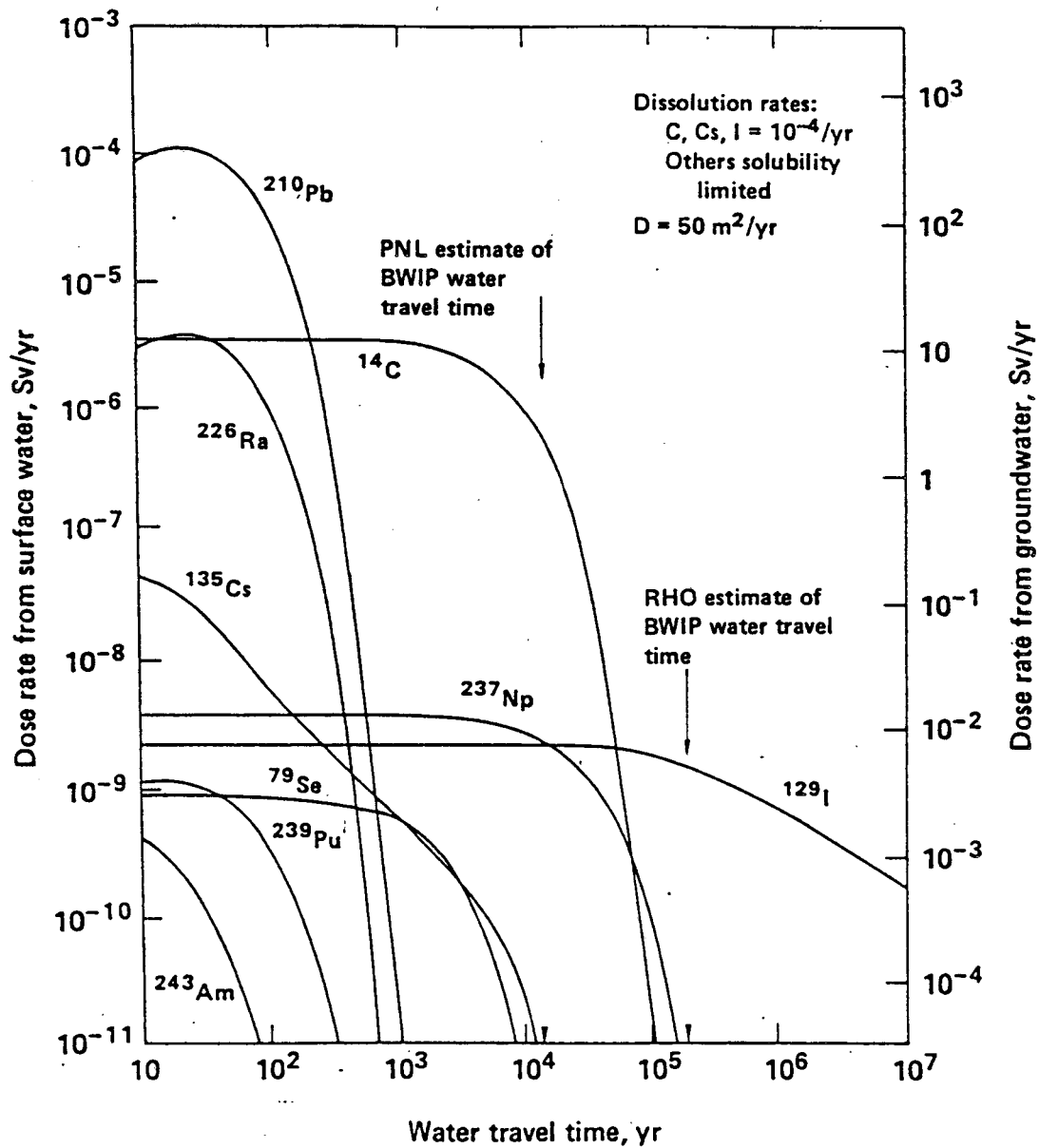


Figure 9. Individual radiation dose as a function of water travel time for basalt, spent fuel, solubility-limited dissolution.

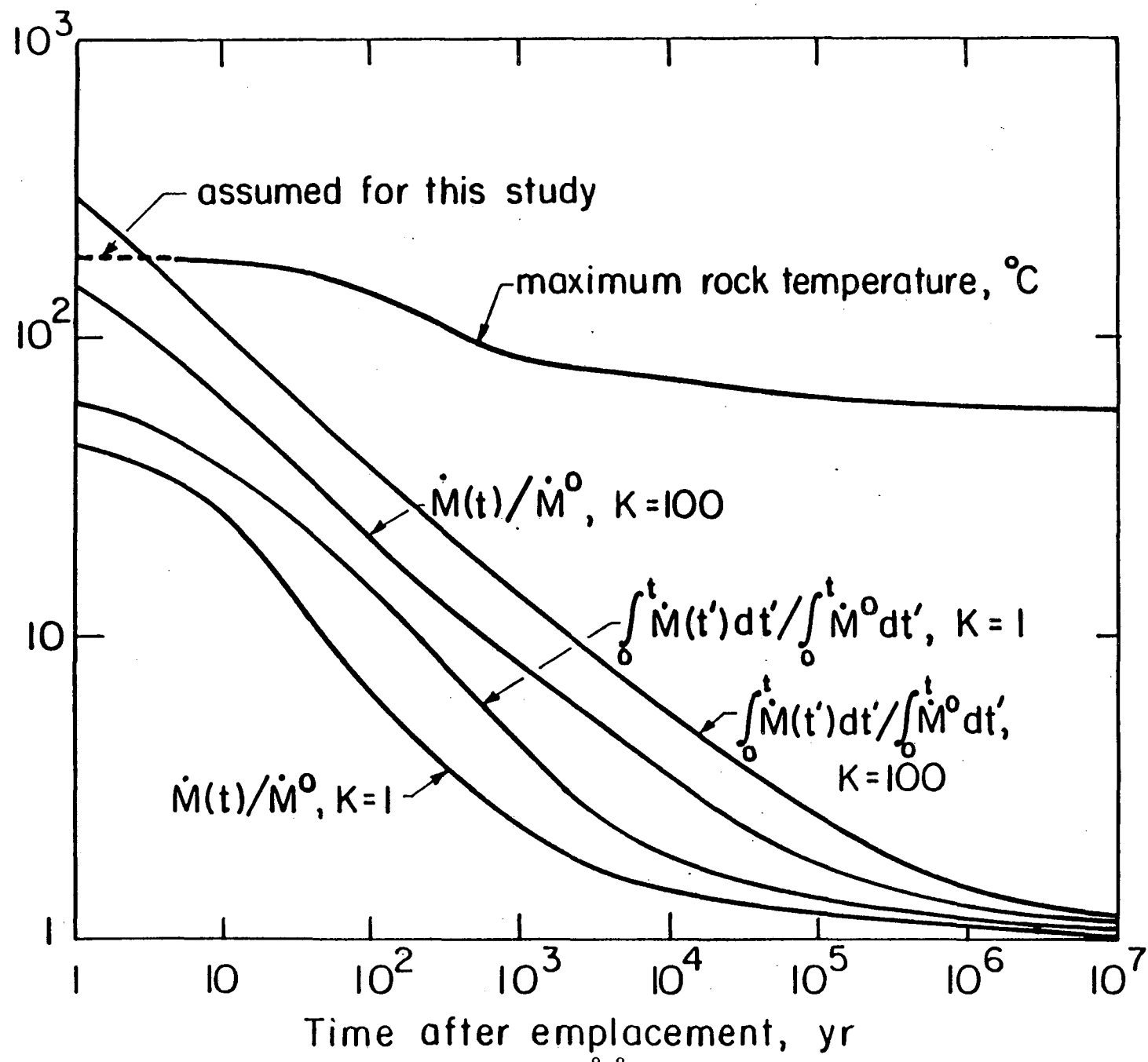


Figure 10. Time-dependent dissolution rate ($\dot{M}^0$ = dissolution rate at ambient steady state).

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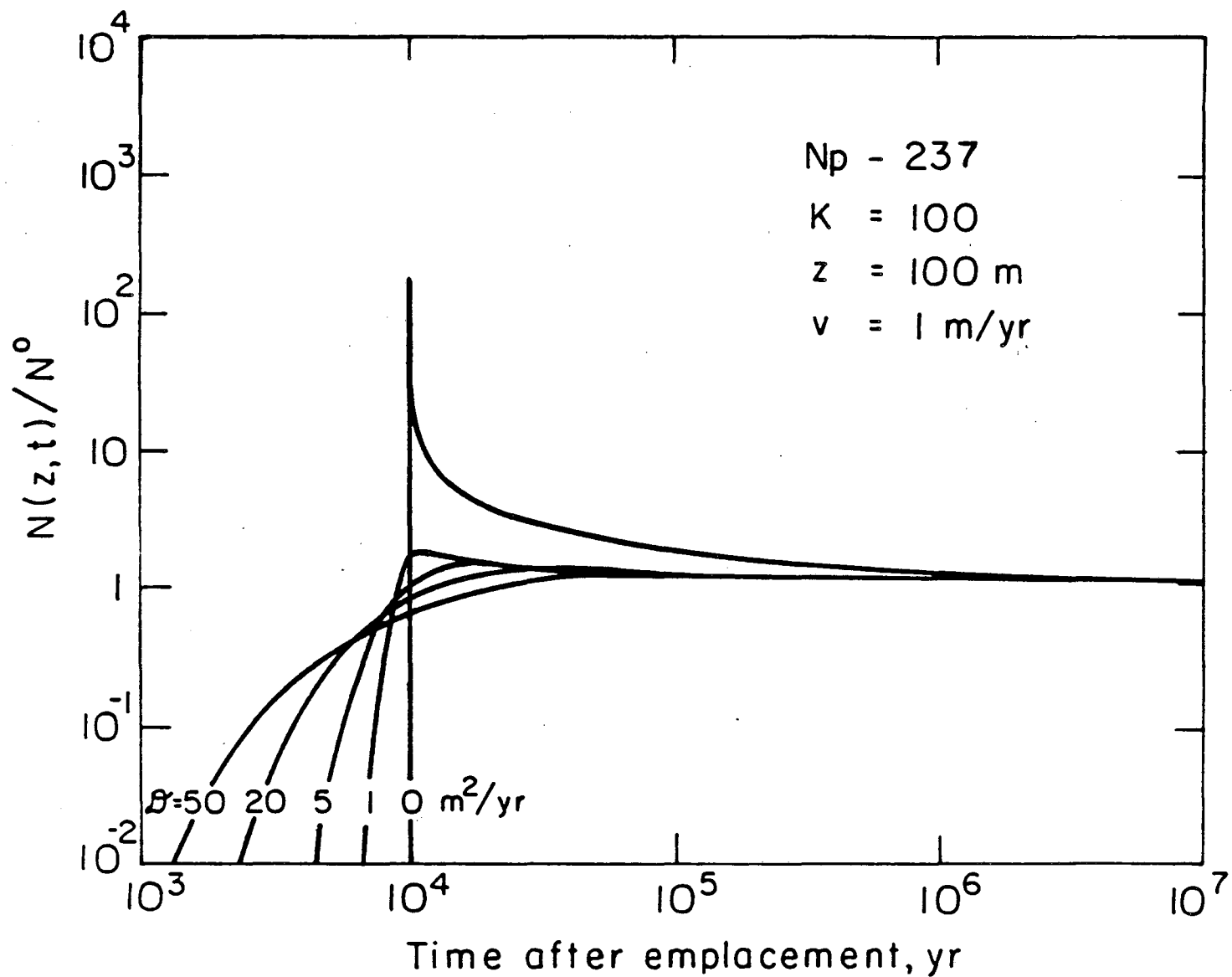


Figure 11. Effect of time-dependent dissolution on the concentration of Np-237 in groundwater.

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