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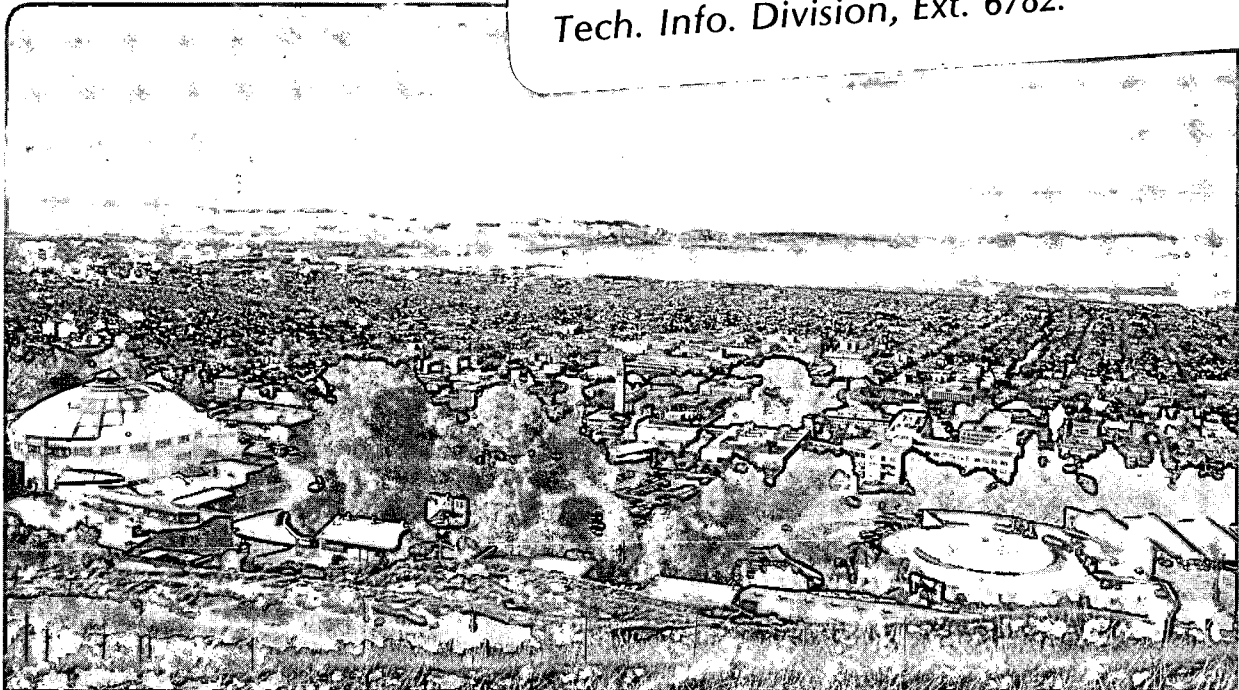Seattle, WA, May 16-20, 1983

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T.H. Pigford

May 1983

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LONG-TERM ENVIRONMENTAL IMPACTS
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Long-Term Environmental Impacts of Geologic Repositories

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1. ABSTRACT

This paper summarizes a study of the long-term
environmental impacts of geologic repositories for radioactive
wastes. Conceptual repositories in basalt, granite, salt, and
tuff were considered. Site-specific hydrological and geochemical
parameters were used wherever possible, supplemented with generic
parameters when necessary. Radiation doses to future maximally
exposed individuals who use the contaminated groundwater and
surface water were calculated and compared with a performance
criterion of 10^{-4} Sv/yr for radiation exposures from probable
events. The major contributors to geologic isolation are the
absence of groundwater if the repository is in natural salt, the
slow dissolution of key radioelements as limited by solubility
and by diffusion and convection in groundwater, long water travel
times from the waste to the environment, and sorption retardation
in the media surrounding the repository. In addition, dilution
by surface water can considerably reduce the radiation exposures
that result from the small fraction of the waste radioactivity
that may ultimately reach the environment. Estimates of
environmental impacts are made both for unreprocessed spent fuel
and for reprocessing wastes. Accelerated dissolution of waste
exposed to groundwater during the period of repository heating is
also considered. This study of environmental impacts is a
portion of a more comprehensive study of geologic waste disposal
carried out by the Waste Isolation System Panel of the U. S.
National Research Council.

2. INTRODUCTION

The Waste Isolation Systems Panel, of the National Research Council, has recently completed a report [1] for the U. S. Department of Energy entitled "A Study of the Isolation System for Geologic Disposal of Radioactive Wastes". Much of that study was concerned with an evaluation of the long-term performance of conceptual waste repositories in various geologic media, and the results are summarized in the present paper. Time-dependent radiation doses to future individuals were calculated [2] for repositories in basalt, granite, tuff, and salt and were compared with the overall performance criterion adopted for this study, expressed as an average annual contribution of 10^{-4} Sv/yr to the lifetime dose commitment for the maximally exposed individual.

The topics dealt with in the present paper are:

- methods of calculation and input data (Section 3)
- individual doses assuming congruent dissolution (Sections 4,5,6)
- corrections due to dispersion (Section 7)
- dissolution rates limited by solubility, diffusion, and convection (Section 8)
- effect of cesium dissolution rate (Section 9).
- individual doses due resulting from solubility-limited dissolution
- radiation doses for unreprocessed spent fuel (Section 11)
- effect of waste dissolution during the period of repository heating (Section 12)
- effect of variations and uncertainties in hydrological and geochemical properties (Section 13)
- conclusions (Section 14)

3. METHODS OF CALCULATION

Calculations of the performance of conceptual geologic repositories were carried out at the University of California at Berkeley [2]. Radionuclide inventories, hydrologic and geochemical parameters and dosimetry factors were supplied by Panel members and consultants, by the U. S. Geological Survey, and by contractors for the U. S. Department of Energy.

3.1. Radionuclide Inventories

Conceptual repositories are assumed to contain radioactive waste resulting from reprocessing 10^5 Mg of uranium fuel from light-water reactors, with radionuclide inventories given in Table 1.

3.2. Dose Conversion Factors

For repositories constructed in rock that normally

contains groundwater, some small amount of radionuclides in the
waste is likely to be released to the biosphere over a long
time. Emphasis is placed here on radiation doses from human
ingestion of radionuclides that dissolve in groundwater and are
transported to the biosphere. For most radionuclides the
radiation doses from direct radiation and from inhalation of
released radionuclides are of lesser consequence.

The maximally exposed individual is one who obtains his
total lifetime intake of potable water from water contaminated by
the released radionuclides and his lifetime consumption of food
from food crops nurtured by the contaminated water. Although the
radiation doses are time-dependent, within an individual lifetime
the rate of release of radionuclides and rate of exposure of such
an individual will be essentially constant, so the individual
dose is calculated on the basis of a 70-yr radiation dose
commitment from radionuclides ingested at constant rate over that
lifetime.

After estimating the time-dependent concentrations of
radionuclides in surface water we obtain the lifetime (70-year)
dose to the maximally exposed individual by using the pathway and
intake calculations of Napier, Kennedy, and Soldat [3], whose
radiation dose factors are derived from the 1959 report of the
International Commission on Radiological Protection (ICRP). We
then adjust the calculated doses for some of the critical
radionuclides by using new calculational techniques and metabolic
data presented in the 1980 report of the ICRP [4], using
correction factors calculated by Runkle and Soldat [5] that
reduce the calculated dose from radium-226 by a factor of 90 and
increase the calculated dose from lead-210 and neptunium-237 by a
factors of 4.4 and 200, respectively. ICRP-30 corrections for
other radionuclides, of lesser importance in the dose calcu-
lations, are not included in this study. The calculated 70-year
dose to the individual is then divided by 70 to obtain an average
yearly dose rate to the individual. The dose conversion factors
thus derived are listed in Table 2.

3.3. Hydrologic and Geochemical Parameters

Hydrologic properties adopted for the calculations are
the flow rates of surface water and of potentially contaminated
groundwater shown in Table 3. The values of solubilities used in
these calculations are the "most probable values" given in Table
4, and the values of retardation coefficients in Table 4 are the
"suitably conservative values", prepared for the Waste Isolation
System Study by Krauskopf [6].

The volumetric flow rate of contaminated groundwater is
the sum of the estimated volumetric flow rates of contaminated
water leaving the repository proper, later diluted by interflows
from aquifers in the surrounding media. This effective flow rate

of contaminated groundwater is expected to increase with distance
from the repository, corresponding to increasing dilution of
dissolved radionuclides. However, no such distance-dependent
data on the flow rates of potentially contaminated groundwater
were available from the DOE and USGS sources for the generic or
proposed repository sites, so a constant value of this flow rate
is adopted for each repository for this study.

Because no groundwater exists within natural salt, the
contaminated flow from a salt repository is assumed to result
from a major diversion of an aquifer in surrounding non-salt
strata, which then dissolves and flows through the salt, dis-
solves waste, and joins an aquifer that then flows eventually to
the surface. The resulting salinity of the aquifer flow will
affect the retardation constants for certain radionuclides,
particularly cesium.

Also shown in Table 3 are volumetric flow rates of water
in rivers that may be typical of flowing surface water into which
the contaminated groundwater discharges. Dilution by the river
flow further reduces the concentrations and resulting individual
radiation doses. Relying here upon effective groundwater travel
times from the repository to the river, as supplied by the
indicated sources of these data, our estimated radiation doses
from river water are not affected by uncertainties in the
effective volumetric flow rate of contaminated groundwater.

The data in Table 3 for water transport time from the
repository to the environment are not used as input for the long-
term release calculations. Instead we adopt the average water
transport time as a variable parameter that depends upon its
location.

3.4. Calculation of Radionuclide Migration

The concentration of each radionuclide in a migrating
decay chain was calculated as a function of distance from the
waste packages and s a function of time since the beginning of
waste dissolution using the exact analytical solutions for one-
dimensional advective transport in a porous medium with
dispersion [15]. At any given distance from the waste packages,
expressed either in a lineal distance or in terms of a
groundwater travel time, the time-dependent concentration is then
searched to obtain the temporal maximum concentration for that
location. It is assumed that radionuclides discharged to a river
mix quickly with the river flow at the discharge location. The
maximum concentration of each radionuclide is multiplied by the
dose-conversion factor from Table 2 to obtain the maximum dose
rate to the individual.

These analytical equations are solved with computer
programs UCB-NE-10.2 and UCB-NE-25, used as benchmark tests for

the international INTRACOIN workshop [16] on methods for
predicting radionuclide migration in geologic media. The effects
of multidimensional dispersion, two-dimensional flow pathways,
and of nonequilibrium transport were examined using the
calculational techniques of Pigford, Chambre' et al. [17].

Calculations of dose rates based upon the assumption of
congruent dissolution are first presented, for comparison with
predictions from the many other published calculations that have
assumed congruent dissolution [11,18,19]. After reviewing the
results for congruent dissolution the importance of later
corrections for solubility-limited diffusive-convective dissolu-
tion will be more readily apparent.

4. TIME-DEPENDENT RADIONUCLIDE DOSES FROM GROUNDWATER TRANSPORT

Figure 1 shows calculated radiation dose to the
individual from different radionuclides as a function of time
after emplacement, for a hypothetical basalt repository loaded
with reprocessing waste, with an assumed water travel time from
waste to the environment of 1000 years, corresponding to a water
travel distance of 1 km at an assumed pore velocity of 1 m/yr.
Dissolution of waste is assumed to begin at the time of
emplacement, and a constant congruent fractional dissolution rate
of all radionuclides of 10^{-4}/yr is assumed. The radiation doses
shown in Figure 1 are only relative doses. The actual radiation
doses depend in magnitude upon the volumetric flow rate of water
carrying the released radionuclides. That is dealt with later.

Because our dose criterion is in terms of the dose to
the maximally exposed individual, we will be concerned with the
maximum dose predicted for each radionuclide. Subsequent
calculations for conceptual repositories will show only the
maximum of each of the radionuclide release bands illustrated in
Figure 1, although for any change in a repository-site parameter,
such as water travel time, the maxima are recalculated.

5. EFFECT OF WATER TRAVEL TIME ON RADIATION DOSES, CONGRUENT DISSOLUTION

5.1. Introduction

We will first examine the peak radiation doses calculated
for congruent dissolution of the waste and neglecting dispersion,
so as to identify the effect of radionuclide inventories,
radioactive decay, water travel time, and sorption delays on the
isolation process. Corrections for dispersion are then
introduced in Section 7, and the effect of more realistic
dissolution rates is considered in Section 8. Dissolution is
assumed to begin shortly after the repository is completely
filled with waste, and all waste is assumed to undergo
dissolution at the same time and rate. The results are

illustrated for a basalt repository, although similar
calculations are presented elsewhere [1,2] for conceptual
repositories in granite, salt, and tuff.

5.2. Basalt

The peak doses from the more important radionuclides,
calculated for various water travel times, are shown in Figure 3
for a basalt repository. The groundwater flow is derived from
data supplied by Brown and Deju [7] for the Basalt Waste
Isolation Project (BWIP), and the river flow rate is that for the
Columbia River, into which groundwater from the potential BWIP
site is predicted eventually to discharge. For assumed
groundwater travel times to the Columbia River between about 10
and 1000 years the principal contributor to the individual dose
is neptunium-237. The calculated dose rate of 1.6×10^{-5} Sv/yr
from river water is sixfold below the performance criterion of
10^{-4} Sv/yr adopted for this study. Carbon-14, assumed not to be
retarded by sorption, is not attenuated appreciably by decay
during this time period and contributes 4×10^{-6} Sv/yr to the
individual dose.

For the average groundwater travel time to the Columbia
River of 1.9×10^5 yr, carbon-14 and most of the other
radionuclides are attenuated by decay before reaching the
environment. For this groundwater travel time the maximum dose
to the individual is due to neptunium-237 and lead-210. The
latter reaches the biosphere by the migration of uranium-238 and
its decay daughters. The calculated dose rate is 6×10^{-9} Sv/yr,
over four orders of magnitude below the performance criterion of
10^{-4} Sv/yr.

For the average groundwater travel time to the Columbia
River of 1.5×10^4 yr estimated by PNL (Dove [13]; Dove et al,
[14]), the maximum dose to the individual is about 10^{-5} Sv/yr
from neptunium-237, tenfold below the performance criterion of
10^{-4} Sv/yr.

Because of the relatively low flow rates of groundwater,
the dose rates to individuals, who theoretically use this
contaminated groundwater for drinking and for irrigation, are all
far above the performance goal of 10^{-4} Sv/yr. In calculating the
doses from using groundwater, a constant groundwater flow rate
[7] has been assumed. This is the effective value after the
groundwater has traveled for 10^4 years from the repository. As
indicated by the basalt data in Table 3, the effective volumetric
flow rate of contaminated groundwater increases with distance
from the repository, and hence increases with water travel time,
due to mixing with interflows. If sufficient data were available
to include this effect, the dose rates from groundwater would
decrease more rapidly with increasing groundwater travel time
than shown in Figure 3.

More realistic estimates of the performance of a basalt repository, and of repositories in other media, are presented in Section 10.

6. EFFECT OF TIME DELAY BEFORE DISSOLUTION

Assuming that all radionuclides undergo congruent dissolution at the same constant fractional release rate, the effect of a time delay before the beginning of dissolution can be deduced directly from the decay equations. A 1000-yr delay decreases the carbon-14 dose by 11 percent. It considerably decreases the dose from strontium-90 and americium-241, which are important only in the very near field. It increases the near-field dose from plutonium-239, neptunium-237, and uranium-234 by allowing time for the decay of their precursors. All of these decay effects are relatively small.

No important effect of delay upon dose rate, with constant fractional dissolution rate, would be expected until the delay is of a time comparable to the half lives of the radionuclides, or of their precursors, that are the important contributors to the radiation doses in the biosphere. Such delays are likely in natural salt, wherein continued dissolution can begin only after the unlikely intrusion of flowing water into the salt.

In Section 12 we analyze the thermal effects on dissolution rates and radiation doses if dissolution begins during the period of repository heating.

7. EFFECT OF DISPERSION

The locally non-uniform distribution of water velocities through pores and fractures can act dispersively on the concentration gradients at the edges of the moving plumes of dissolved radionuclides, resulting in broadening of the concentration bands and some reduction in concentrations. The most important effect on calculations of the rate of discharge of radionuclides to river water, and on the maximum individual dose therefrom, is that resulting from dispersion in the direction of flow, i.e., axial dispersion. Little is known about the proper value of the dispersion coefficients for the media considered here, but a value of 50 m^2/yr for the axial dispersion coefficient is assumed for the purpose of this study [7].

Dispersion corrections to the maximum dose rates from carbon-14, cesium-135, iodine-129, neptunium-237, and radium-226 for a basalt repository are shown in Figure 4. As in the case of strongly sorbing cesium, dispersion reduces the maximum dose as the radionuclide travel time increases. Attenuation is greater

when the radionuclide travel time is much greater than the dissolution time of the waste.

        Groundwater flowing past an individual package of dissolving waste creates a plume of contaminated water downstream of each waste package. Transverse dispersion soon merges the plumes from the many waste packages into a broad plume of essentially uniform concentration [17,21], that may later become diluted with merging interflows through contacts after it leaves the host rock of the repository. This local near-field dispersion does not affect the maximum far-field doses calculated in this study.

8. DISSOLUTION RATES LIMITED BY SOLUBILITY, DIFFUSION, AND CONVECTION

8.1. Summary of Solubility-Limited Dissolution Theory

        It is not necessary or realistic to assume that all radionuclides dissolve congruently with the same fractional release rate, as is usually assumed in other analyses of repository performance [11,18,19]. Congruent dissolution of most of the radionuclides in the waste is not expected to occur. Many of the important radioelements are so insoluble that they cannot go into solution as rapidly as the waste matrix itself.

        Other studies [22,23,24] have treated 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 diffusional resistance in the concentration boundary layer adjacent to the waste surface, and it assumes instantaneous diffusive and dispersive transport 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.

        The analyses by Chambre' et al. [21,25,26] 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 [27], 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 (Altenhein, et al. [28],

Malow [29], Wicks, et al. [30]) of actinides and other low-solubility constituents in precipitates formed on borosilicate-glass surfaces during laboratory leaching experiments.

Assuming that the waste form is in contact with porous rock or with backfill of essentially 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 5. 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 [1,2].

8.2. Mass Transfer From The Waste by Diffusion and Convection

The diffusion-and-flow calculation 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 our estimates we assume that the diffusion coefficient is equal to that in a free liquid, conservatively disregarding the tortuosity of the pores [31].

The fractional release rate f_j of the elemental species j, and of its isotopes, is calculated [21,25] at steady state to be:

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

where N_j^* is the solubility-limited 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.

Table 5 gives values of the solubility of silica and the solubilities in water of radioelements in borosilicate glass. The solubilities are those from Table 4. Also listed in Table 5 are the bulk densities and the calculated fractional release rates for a typical waste form of glass exposed to ground water at an approach velocity of 1 m/yr in a rock of one percent

equivalent porosity. The assumed diffusion coefficient of 10^{-5} cm^2/sec is typical for an electrolyte in water. Tortuosity in granite can result in a 100-fold to a 1000-fold reduction in D_j [34,35].

In some repositories the groundwater velocity is estimated to be so low, even during the thermal period [7], that Equation (1) is not applicable. A separate equation, applicable in the limit of very small velocities, has been derived by Chambre' [21,16]. The resulting fractional release rates are only fourfold below those in Table 5. Equations for the transient solubility-limited dissolution rate, before the long-term steady state is reached, have also been derived [21,26].

8.3. Comparison With Laboratory Leach Data

Table 5 also gives values of the fractional release rates for silica and for various radioactive elements, calculated from experimental values of leach rates reported from laboratory experiments [33]. Note that, for substances having limited solubility, the values of f_j computed from Equation (1) are smaller then several values derived from the laboratory leach tests. One must conclude, therefore, that for slightly soluble species in waste which has been embedded in a repository rock and is surrounded by Darcy flow the slow diffusion and slow movement of the liquid around the waste containers may be more significant in controlling the net rate of dissolution than the rate at which the substances inside the waste material reach the surface of the waste form. If the solubility is very small, the rate of escape into ground water will be determined primarily by the properties of the porous rock and the velocity of ground water; if the solubility is sufficiently large the kinetics of the interaction between the solid waste constituents and water may dominate.

8.4. Application to a Repository

Predicted solubility-limited fractional dissolution rates in Table 5 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 means that 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 possibly for 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.

The above analyses 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 are given in Table 4, and the effects of solubility uncertainties on repository performance are discussed in Section 13.

8.5. The Effect of Colloids

For such low-solubility species it is possible that the rate of release 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 [36]. Otherwise, colloidal particles may be trapped in the pores and not move [37, 38,39].

8.6. The Need for Verification

Although the diffusive-convective equations for dissolution rate contain no unknown constants to be emperically determined, other than the well-defined physico-chemical properties of solubility and diffusion coefficient, these predictive 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' [21,26]. For a non-sorbing species 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 is 310 yr for the parameters given in Table 5. The time is much shorter, of the order of a few years, when the flow velocity is great enough to warrant use of Eq. (1). For a sorbing species, with a retardation coefficient greater than unity, the equilibration time is proportional to the retardation coefficient of that species 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.

8.6. Dissolution of Carbon-14 and Iodine-129 Wastes

Carbon-14 is not a constituent within the high-level waste packages, because it is separated in fuel reprocessing. However, assuming that carbon-14 is recovered separately and is emplaced in a geologic repository, we can estimate the solubility-limited dissolution rate of a hypothetical waste package containing carbon-14. It is assumed here that all of the carbon-14 recovered from reprocessing the entire 10^5 Mg of uranium fuel, whose waste represents the full loading of a repository, is accumulated and incorporated into a single waste package. The total inventory of carbon-14 is 19.8 kg. We assume that the carbon-14 is in the form of granulated calcium carbonate, with a total mass of 217 kg. The heat generation rate within this special carbon-14 package is about 40 watts, far less than for a single high-level waste package. Therefore, the carbon-14 package can be assumed to operate at the ambient temperature.

At $25^\circ C$ the solubility of calcium carbonate is 1.4×10^{-5} g/cm^3 [40]. Assuming that the carbon-14 waste package is of the same overall dimensions as the high-level waste package assumed in Table 5, we estimate a solubility-limited fractional release rate of carbon-14 of 4×10^{-7}/yr. This reduces the individual dose rate from carbon-14 by a factor of 500 below that shown in Figures 3 for congruent dissolution.

Similarly, separated radioiodine could be reacted with silver to form low-solubility silver iodide, with a predicted fractional dissolution rate of 8×10^{-11}/yr [1,2]. However, excess halide or ammonium ions in the groundwater could solubilize and complex the iodine.

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

9. CESIUM DISSOLUTION RATE

All of 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 may be 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 5 to be 1.1×10^{-6}/yr, if cesium dissolves congruently with the silica matrix. In Figure 6 we show the effect on 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 up to the value of 1.6×10^{-3}/yr inferred from laboratory experiments on borosilicate glass [41]. With dispersion, cesium-135 is important only within the near-field of the repository. The three dispersion curves coalesce at water travel times greater than about 10^3 yr. This illustrates the important conclusion that the far-field peak concentration of a radionuclide migrating under the influence of dispersion can be independent of the dissolution rate of that radionuclide, provided that the migration time is much longer than the dissolution time [2]. This insensitivity of cesium dose to cesium dissolution rate is important for repositories in basalt, granite, or tuff, because of the relatively high cesium retardation and consequently high dispersion during the water travel times of interest. Dissolution rate is more important for more weakly sorbing radionuclides, and it is more important for cesium released from a salt repository, where sorption and dispersion of cesium are less effective.

10. EFFECT OF WATER TRAVEL TIME ON RADIATION DOSES, SOLUBILITY-LIMITED DISSOLUTION

10.1. Basalt

The solubility-limited dissolution rates of Table 5 are applied individually to the radionuclides to calculate the peak individual doses for the basalt repository. The results shown in Figure 7 are calculated for solubility-limited dissolution of all radionuclides except cesium-135. The effects of axial dispersion, with an assumed dispersion coefficient of 50 m^2/yr, are including in these calculations of doses from repositories with solubility-limited dissolution.

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 daughter radium-226 and lead-210. Based upon these predictions, and based upon the estimate by the BWIP project [7] of a water travel time to the environment of 1.5×10^5 yr, 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 [13] and Dove et al. [14], the maximum dose to the individual will be about 3×10^{-9} Sv/yr, over four orders of magnitude below our individual-dose criterion.

Figure 7 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 with regard to 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 actually occur.
There are also many uncertainties and other phenomena, not taken
into account in the present analysis, that must be determined
before the margin of uncertainty in the predicted dose can be
determined. Such analysis is beyond the scope of this study.
However, it appears that the large margin between the calculated
dose in river water and the goal of 10^{-4} Sv/yr may sufficiently
allow for these uncertainties.

10.2. Granite

The solubility-limited dissolution rates of Table 5 are
adopted here for the granite repository, and the predicted doses
are shown in Figure 8. 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 flowrate of river water 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 as long as 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.

10.3. Salt

The solubility-limited dissolution rates of Table 5 are
adopted for a salt repository, although it might be expected that
the geochemical environment in the media adjacent to a salt
repository could result in somewhat greater solubilities and
dissolution rates than for basalt and granite. The estimated
radiation doses for a salt repository are shown in Figure 9.
Here cesium-135 dominates the dose for water travel times greater
than 10^5 yr. Determination of the appropriate dissolution rate
for cesium-135 is of special importance for a salt repository.
For the cesium retardation coefficient adopted here, cesium
dispersion is not sufficient to result in the insensitivity of
predicted doses to the dissolution rate of cesium in a salt
repository.

10.4. Tuff

Adopting the solubility-limited dissolution rates of
Table 5, the calculated doses for a tuff repository are shown in

Figure 10. 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. We have no estimates of groundwater flowrate for a tuff site, so we have assumed the same flow rate of contaminated groundwater that has been assumed elsewhere for generic sites in other media [11]. Because there is no flowing surface water into which potentially contaminated groundwater from the tuff site can discharge, the calculations are limited to the doses from direct use of the contaminated groundwater. Data in Table 3 indicate that groundwater is already being used in this vicinity, and attention must be focused on possible future use of the potentially contaminated groundwater by humans, for potable water and irrigation.

Tentatively adopting 4300 yr as the possible water travel time from a repository in saturated tuff to an off-site well, the radiation dose will be dominated by carbon-14 or, depending upon its dispersion coefficient, by cesium-135. Adopting 2.1×10^4 yr as the possible water travel time to the same off-site location from a repository in unsaturated tuff, the carbon-14 dose is 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, but the generally arid area of the tuff site suggests that the potentially contaminated groundwater is likely to be used.

This performance analysis of a tuff repository illustrates one of the differences between adopting a performance criterion in terms of an individual dose or in terms of a population dose or radioactivity release limit, the latter appearing in EPA's proposed standard [42]. Our calculations indicate that this possible tuff repository would meet EPA's proposed 10,000-year release limits at the EPA-NRC 10-km location, assuming the numerical criteria in NRC's proposed 10 CFR 60 regulation [43] and assuming that those numerical criteria can be met by all of the radionuclides with releases above NRC's 0.1 percent threshold. Yet, with the parameters adopted for the calculations herein, a tuff repository may not meet the individual dose rate criterion. EPA's proposed standard and the NRC staff's proposed numerical criteria are not concerned with the effects of water volumetric flowrates, and they are not concerned with concentrations and individual doses from radionuclides in groundwater and surface water.

11. RADIATION DOSES FOR UNREPROCESSED SPENT FUEL

The calculated radiation doses for a basalt repository completely loaded with unreprocessed spent fuel are shown in Figure 11 for solubility-limited dissolution. Comparing with the similar data in Figure 7 for a basalt repository loaded with reprocessing waste, the mass dissolution rate of solubility-limited parent nuclides are the same as for reprocessing waste. The calculated doses from neptunium-237 are the same, and 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. In the field near the waste packages the higher initial uranium-234 of the spent fuel creates more thorium-230 and much greater concentrations of radium-226, for water travel times less than about a thousand years.

Assuming that the dissolution rate of cesium is the same for spent fuel as for reprocessing 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 9, should make the cesium dose rate for spent fuel insensitive to release 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 package surface area to the inventory of each of these two radionuclides in each package results in no solubility limits on the release of these radionuclides. Their dissolution rates are likely to be determine 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 relatively 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 separated radionuclides.

The differences between spent fuel and reprocessing waste, as illustrated here for a basalt repository, will apply as well to the other repository media. For the reasons outlined above, the reprocessing of spent fuel allows preparation of waste forms that will improve repository performance.

12. EFFECT OF REPOSITORY HEATING

Although the U. S. Nuclear Regulatory Commission [44,45] has concluded that temperature effects are important and should be avoided by a 1000-year overpack, no calculations or experiments were available to justify these conclusions. In the absence of any 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 be increased because the higher temperatures increase both the solubilities and the diffusion coefficient in the liquid boundary layer. Using data supplied by the BWIP Project [7] for time-dependent temperatures and groundwater flowrates during the thermal period, we have estimated [1,2] the effects of these parameters on the dissolution rate of exposed borosilicate glass, approximating the dissolution rate from the steady-state dissolution rate equations of Chambre' et al. [21,26]. From these results, exposure of the waste form to the groundwater during the thermal period at the maximum 5-year temperature of 250°C could result in a silica dissolution rate about forty times greater that the rate at the ambient temperature of 57°C, or about sixty times greater than the dissolution rate at 20°C listed in Table 5. After 100 years, as the rock temperature at the emplacement hole decreases, the silica dissolution rate decreases to about sevenfold greater than the ambient value.

As a first approximation, we assume that the same proportional increase applies to all of the low-solubility radioelements, with the consequence that exposure of all the waste during the thermal period could cause a fortyfold increase in the calculated near-field doses, where the main effect is the greater dissolution rate resulting from greater solubilities and diffusion rates. For the low water velocities in the basalt host rock, thermally affected flow velocities have little effect upon the dissolution rate, because at these low velocities transport through the liquid boundary layer is controlled by molecular diffusion, and transport through the boundary layer by convection is relatively unimportant.

The effect of repository heating upon the doses from radionuclides reaching surface water is not so great, because of dispersion during transport. If the temperature were to cause a fortyfold greater dissolution rate for a few hundred years, there would be a fortyfold increase in the concentration of each radionuclide at the leading edge of its chromatographic band in the field near the repository. This is a rather short-duration increase in the near-field concentration for the time periods of hydrogeologic transport. Because it occurs during a time interval much smaller than the radionuclide travel time to the

biosphere, the concentration spike will almost completely
disappear because of dispersion by the time it reaches the
biosphere. Thus, what can be an appreciable concentration
increase in the near field of the repository may result in a
rather small effect when it reaches the biosphere.

These calculations suggest that the performance of the
repository with waste exposed during the thermal period is
amenable to calculation, provided data are obtained on
temperature-dependent solubilities. They show that the effect of
thermally-induced flow upon the dissolution rate may not be
important in a repository wherein the flow velocities are
normally quite low. They show that appreciable concentration
increases can occur in the near field of the repository during
the thermal period, but that for long water transport times to
the biosphere the concentration increases may be relatively
small. Although there are many uncertainties in these estimates,
it may be easier and more cost effective to clear up these
uncertainties by careful experiments and analyses, especially on
temperature-dependent solubilities, than to provide the 1000-yr
corrosion resistant overpack proposed by the U. S. Nuclear
Regulatory Commission [43,44,45] to reduce the uncertainties in
estimating repository performance during the thermal period.

13. EFFECT OF VARIATIONS AND UNCERTAINTIES IN HYDROLOGICAL
     AND GEOCHEMICAL PROPERTIES

Figures 7-11 show that radiation doses are relatively
insensitive to water travel time, for travel times in the range
of a few hundred to a few thousand years for basalt, granite, and
tuff, and up to about 10^5 yr for a salt site. The greatest
reduction in dose occurs when water travel time is increased to
about 10^5 yr for basalt, granite, and tuff and to about 10^6 yr
for a salt site. Elsewhere [1,2] we present estimates of the
effects of uncertainties in hydrological and geochemical
parameters on the predicted performance of repositories. The
calculations rely in part upon the range of solubilities and
retardation constants estimated by Krauskopf [1,6] in Table 4.
The effect of these uncertainties on the predicted individual
dose from neptunium-237 are illustrated in Figure 12.

A major uncertainty, not subject yet to calculation, is
the possibility of climatic changes that could occur over the
hundreds of thousands of years that have been considered in this
analysis.

14. CONCLUSIONS

Among the several conclusions reached by the Waste
Isolation Systems Panel [1], selected conclusions relevant to the

relevant to the analysis of the long-term environmental
performance of conceptual repositories are:

o   Timely development of geologic waste-isolation systems
and assessment of their adequacy require a general criterion that
defines acceptable overall performance. This criterion should
include a numerical criterion for the lifetime radiation dose to
the maximally exposed individual.

o   When buried waste solids are exposed to groundwater,
most of the radionuclides can be contained within the waste
package or within the geologic media, or both, long enough to
disappear by radioactive decay; but even after delays of
thousands to millions of years a small proportion of the
radionuclides will still reach the biosphere by groundwater
transport.

o   When groundwater is not initially present in the host
rock of the repository, as in the case of a properly chosen salt
deposit, containment within the waste package is complete unless
humans intrude or unless groundwater intrudes into and flows
through the repository.

o   Laboratory data for the release of radionuclides from
candidate waste forms have not been shown to be applicable to
predicting performance of waste packages in geologic
repositories.

o   Borosilicate glass is the appropriate choice for
further testing and for use in current repository designs.
Although there are ucertainties in its performance in a waste
package during the period of repository heating, its performance
during the post-thermal period remains the most important issue
for geologic repositories.

o   Uncertainties about the physical integrity of
borosilicate glass exposed to leaching solutions at high
temperatures may require that glass high-level waste be protected
from groundwater by a corrosion-resistant overpack when the
repository rock is at elevated temperatures. However, present
performance analyses indicate that such protection may be neither
necessary nor beneficial.

o   Repositories are likely to be loaded initially with
wastes derived from accumulated and aged discharged reactor fuel.
With such loadings, maximum rock temperatures in current
repository designs are predicted to be low enough to warrant
confidence in borosilicate-glass for these initial emplacements.
This provides more time for resolving the performance of waste
forms at the higher temperatures associated with later loadings
in the repository.

o   A continuing back-up program of research and
development on alternative waste forms is recommended.

o   The diffusion-convection theory predicts very low
dissolution rates for low-solubility radionuclides. The
concentrations of these radionuclides later released to surface
water are calculated to be low enough that the predicted doses to
the maximally exposed individuals who utilize this surface water

can be several orders of magnitude below the dose rate criterion
of 10^{-4} Sv/yr adopted for this study.

o   For water travel times of about 100,000 years or
longer, fuel-reprocessing waste can be adequately contained in
granite, basalt, or tuff. Even if water intrudes into a salt
repository within a few hundred thousand years after waste
emplacement, water travel times of 100,000 years or longer after
intrusion will ensure adequate containment of all the
radionuclides in fuel-reprocessing waste.

o   Dilution by surface water to reduce radiation doses to
exposed individuals is important for a repository in basalt at
Hanford, Washington, and for the generic granite repository
assumed for this study, but such dilution is unavailable for a
repository in the tuff site.

o   A repository in unsaturated tuff is expected to provide
much longer time delays for potentially contaminated groundwater
to travel to a given off-site location than is a repository in
saturated tuff.

o   All radionuclides in unreprocessed spent fuel can be
adequately contained. The peak doses from spent fuel, due to
carbon-14 and lead-210, are considerably greater than those from
fuel-reprocessing waste.

o   For all of the host-rock media considered herein,
including a salt site with assumed intrusion of water, the
assumed use of the groundwater for drinking and irrigation before
it mixes with surface water could result in radiation doses that
are greater than the performance criterion of 10^{-4} Sv/yr. For a
repository site located in a region with no flowing surface
water, there will be a greater incentive for future generations
to use the groundwater for drinking and irrigation than in the
case of a site located near a large amount of surface water.

o   Throughout the lifetime of the program to develop and
implement geologic disposal or radioactive waste, programmatic
priorities and decisions must be guided by the predictions of
long-term performance of the geologic disposal system. Greater
emphasis should be given to the development, validation, and use
of techniques for predicting long-term performance.

o   The accuracy of predicting future environmental
releases from a geologic repository decreases as the future time
at which the radionuclides are predicted to reach the environment
increases. However, predictions should be made for all future
times until potentially significant radiation doses are no longer
predicted to occur. Uncertainties in the time-dependent
predictions should be estimated.

o   Uncertainties in predictive accuracy arise principally
from uncertainties in water travel time to the environment, in
volumetric flow rates of potentially contaminated surface water
and groundwater, in solubilities and retardation properties of
key radioelements, and in the calculation of radiation doses that
could result from the use of contaminated groundwater for growing
food and drinking. The latter uncertainties are greater when
estimating population doses than when estimating individual
doses.

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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	$\leqslant 6.5 \times 10^4$	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 Average Lifetime Dose Rate per Unit Concentration
of Radionuclides in Water

Radionuclide	Half-Life(yr)	Average Dose Rate per Unit Concentration[a] $(Sv\text{-}m^3/Bq\text{-}yr)$	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 3 Hydrologic Parameters for Reference Repository Sites

	Repository Rock						
						Tuff[e]	
	Basalt[a]		Bedded[b] Salt	Domal[c] Salt	Granite[d]	Saturated	Unsaturated
Location	Hanford, Wash.		Permian & Para-dox basins	Generic	Generic	Nevada Test Site	
Flow rate through repository (m^3/yr)	17		40 to 400				
Flow rate of contaminated aquifer (m^3/yr)	3.2×10^4 (at 10^4 yr)		1.3×10^3	5.2×10^5			
Flow rate of surface water (m^3/yr)	1.1×10^{11}		3.2×10^8	2.8×10^9	3.8×10^9		
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		1		
Maximum	80	16	10^3		10^2		
Mean	70	14	10^2		10		
Water travel time to biosphere(yr)						1.2×10^3 (to well J13) 4.3×10^3 (to Lathrop Wells)	2.2×10^4 (to well J13) 2.5×10^4 (to Lathrop Wells)
Minimum	1.3×10^4	1.3×10^4	1×10^4		10^3		
Maximum	1×10^6	1.7×10^4	1×10^6		$>10^6$		
Mean	1.9×10^5	1.5×10^4	1×10^5	4×10^4	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 5×10^{-2}		1.8×10^{-1}	0.10	10^{-5} to 3×10^{-2}	0.18(to Lathrop Wells)	

[a] D. J. Brown and R. A. Deju[7].
[b] S. Goldsmith[8], except flow rate of contaminated aquifer is from G. E. Raines[9]. Data assume a salt repository breached by flowing water.
[c] Bechtel Group[10]. Data assume a salt repository breached by flowing water.
[d] Data from[1], except flow rate of contaminated aquifer is from Cloninger and Cole[11].
[e] Tyler[12].
[f] Dove[13], Dove et al.[14].

Table 4 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 / *50* / 200	5 / *50* / 200	5 / *50* / 200	5 / *50* / 200	20 / *200* / 1,000
Sr	High	-0.2	High	-0.2	High	10 / *200* / 2,000	50 / *200* / 2,000	20 / *200* / 10,000	50 / *200* / 5,000	1 / *10* / 100
Zr	-4	-4	-6	-4	-6	500 / *5,000* / 30,000	500 / *5,000* / 10,000	500 / *5,000* / 10,000	500 / *5,000* / 50,000	300 / *1,000* / 5,000
Tc	-3	-10	High	High	High	1 / *5* / 40	1 / *5* / 100	1 / *5* / 100	1 / *5* / 20	1 / *5* / 20
Sn	.3(?)	-4	-4	-4	-4	100 / *1,000* / 5,000	100 / *1,000* / 5,000	200 / *1,000* / 5,000	200 / *1,000* / 5,000	10 / *100* / 1,000
Sb	-3(?)	----	----	----	----	10 / *100* / 1,000	10 / *100* / 1,000	10 / *100* / 1,000	10 / *100* / 1,000	5 / *50* / 500
I	High	High	High	High	High	1 / *1* / 1	1 / *1* / 50	1 / *1* / 1	1 / *1* / 1	1 / *1* / 1
Cs	High	High	High	High	High	100 / *1,000* / 10,000	100 / *1,000* / 10,000	60 / *500* / 10,000	200 / *1,000* / 20,000	1 / *10* / 2,000
Pb	-1	-1	0	-1	0	10 / *50* / 200	20 / *50* / 500	20 / *50* / 500	20 / *50* / 500	5 / *20* / 100
Ra	-2	-3	-1	3	-1	50 / *500* / 5,000	50 / *500* / 5,000	50 / *500* / 5,000	50 / *500* / 5,000	5 / *50* / 500
Th	-3	-4	-4	-4	-4	500 / *5,000* / 10,000	500 / *5,000* / 10,000	500 / *5,000* / 10,000	500 / *5,000* / 50,000	300 / *1,000* / 5,000
U	-3	-3	-5	High	High	10 / *50* / 500	20 / *50* / 1,000	5 / *40* / 200	50 / *200* / 5,000	10 / *20* / 60
Np	-3	-4	-4	-2	-1	10 / *100* / 500	10 / *100* / 500	10 / *100* / 500	10 / *100* / 400	10 / *50* / 300
Pu	-3	-5	-4	-5	-3	10 / *200* / 5,000	100 / *500* / 5,000	50 / *200* / 5,000	500 / *1,000* / 20,000	10 / *200* / 10,000
Am	-4	-8	-5	-8	-5	500 / *3,000* / 50,000	60 / *500* / 50,000	300 / *1,000* / 50,000	200 / *800* / 50,000	300 / *1,000* / 5,000
Cm	-3(?)	----	----	----	----	200 / *2,000* / 10,000	100 / *500* / 10,000	100 / *500* / 10,000	200 / *2,000* / 20,000	200 / *1,000* / 3,000

a/ From Krauskopf [1,5].

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

Waste cylinder: radius = 0.152 m, length = 2.46 m, fission-pruduct 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 SiO$_2$ [32]. Other solubilities are from Table 4.

b/ Calculated from Equation (1).

c/ Data of McVay, et al. [33] for IAEA-type leach tests, with periodic replacement of leachant.
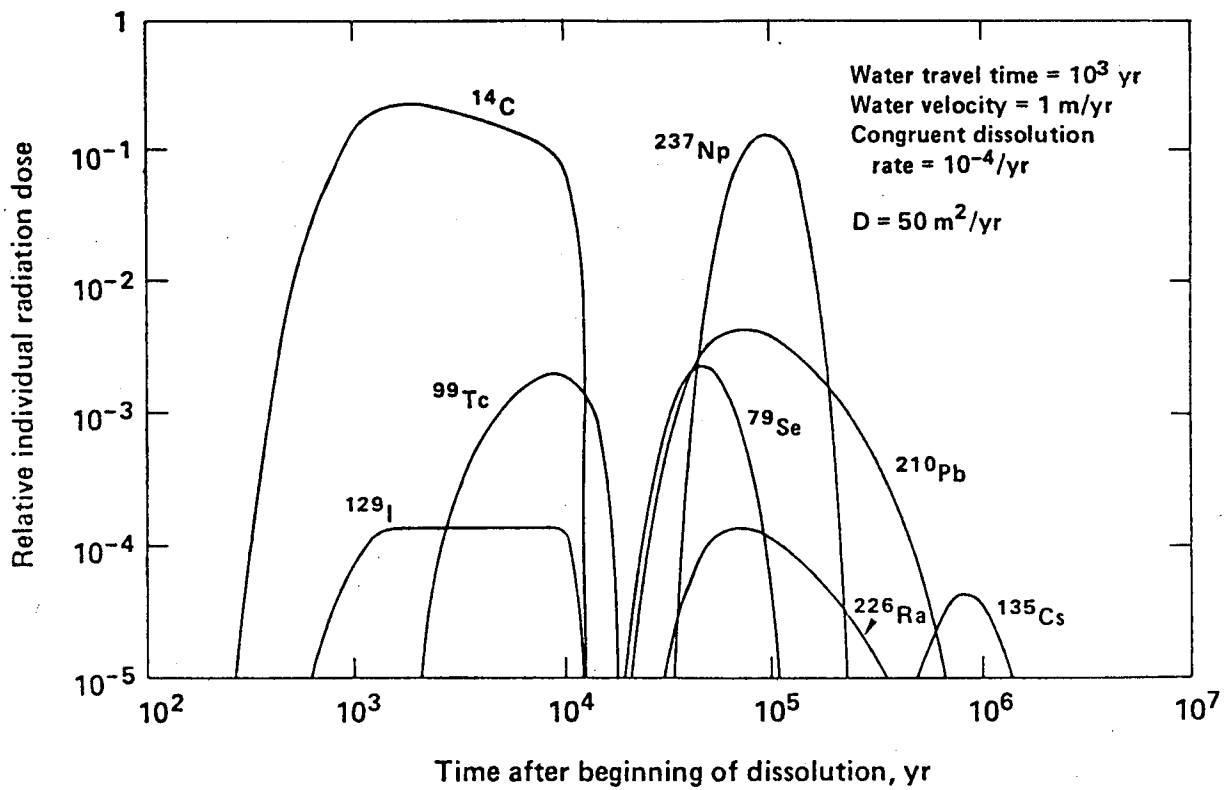
Figure 1. Relative individual radiation dose as a function of time for basalt, reprocessing waste, congruent dissolution

$^{242}\text{Am} \xrightarrow[\text{152 yr}]{\beta^-} {}^{242}\text{Cm} \xrightarrow[\text{163 d}]{\alpha} {}^{238}\text{Pu}$

$\downarrow \alpha \quad \text{86 yr}$

$^{238}\text{U} \xrightarrow[4.51 \times 10^9 \text{ yr}]{\alpha,\ 2\beta^-} {}^{234}\text{U} \xrightarrow[2.47 \times 10^5 \text{ yr}]{\alpha} {}^{230}\text{Th} \xrightarrow[8 \times 10^4 \text{ yr}]{\alpha} {}^{226}\text{Ra} \xrightarrow[1{,}602 \text{ yr}]{\alpha}$
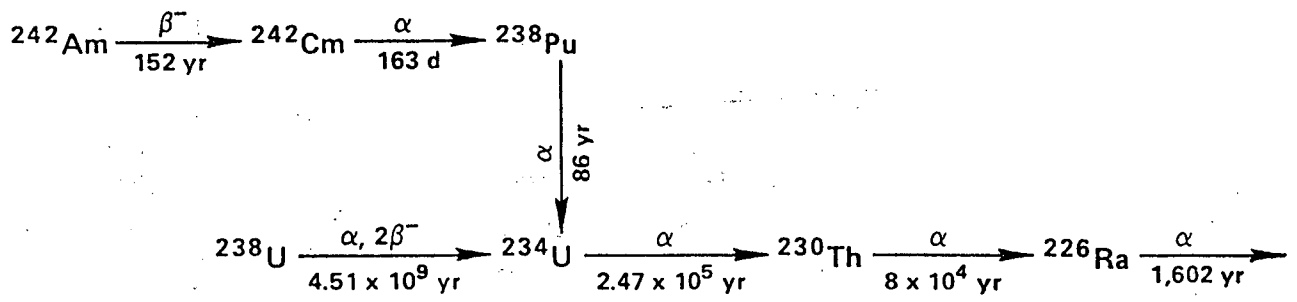
Figure 2. Decay chains leading to radium-226
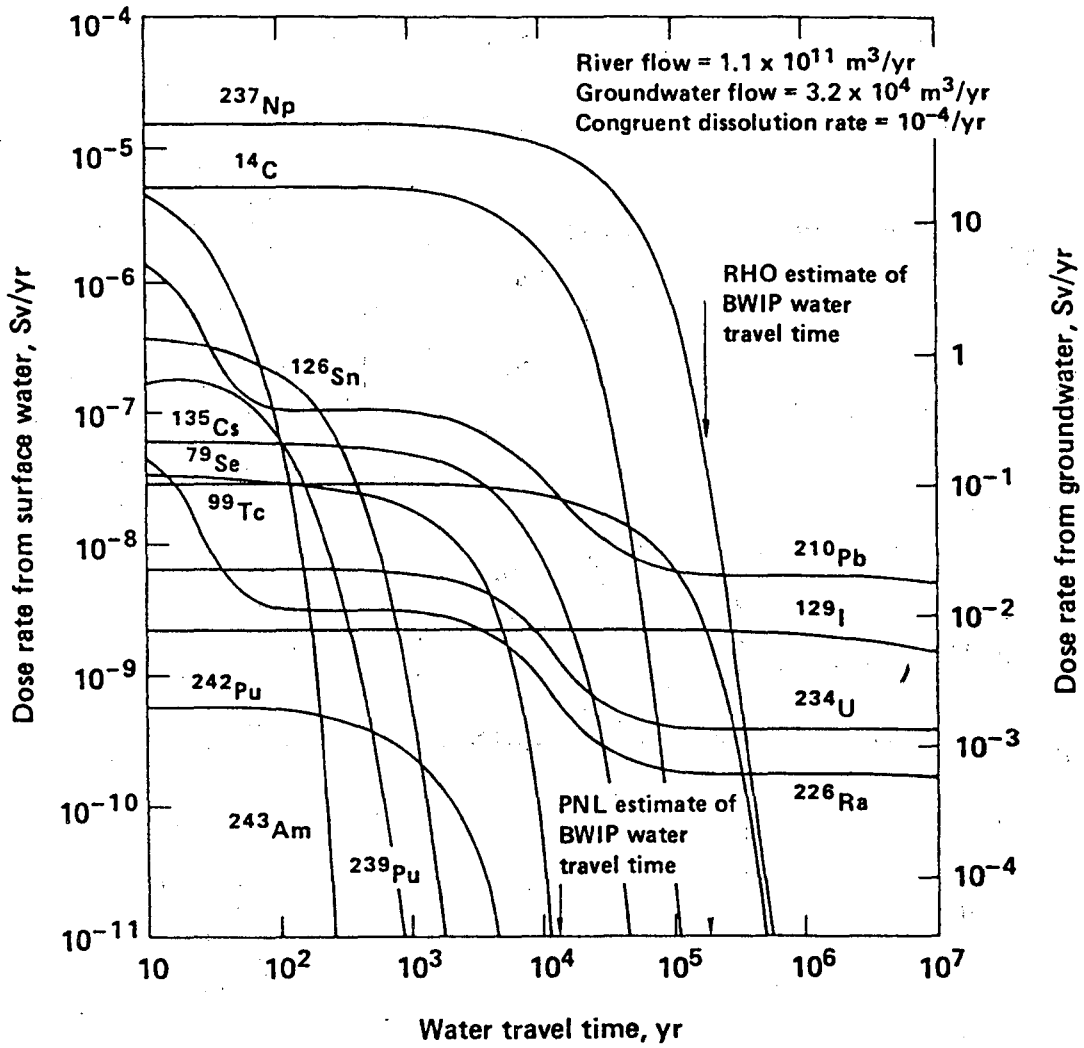
Figure 3. Individual radiation dose as a function of
water travel time for basalt, reprocessing
waste, congruent dissolution, no dispersion.
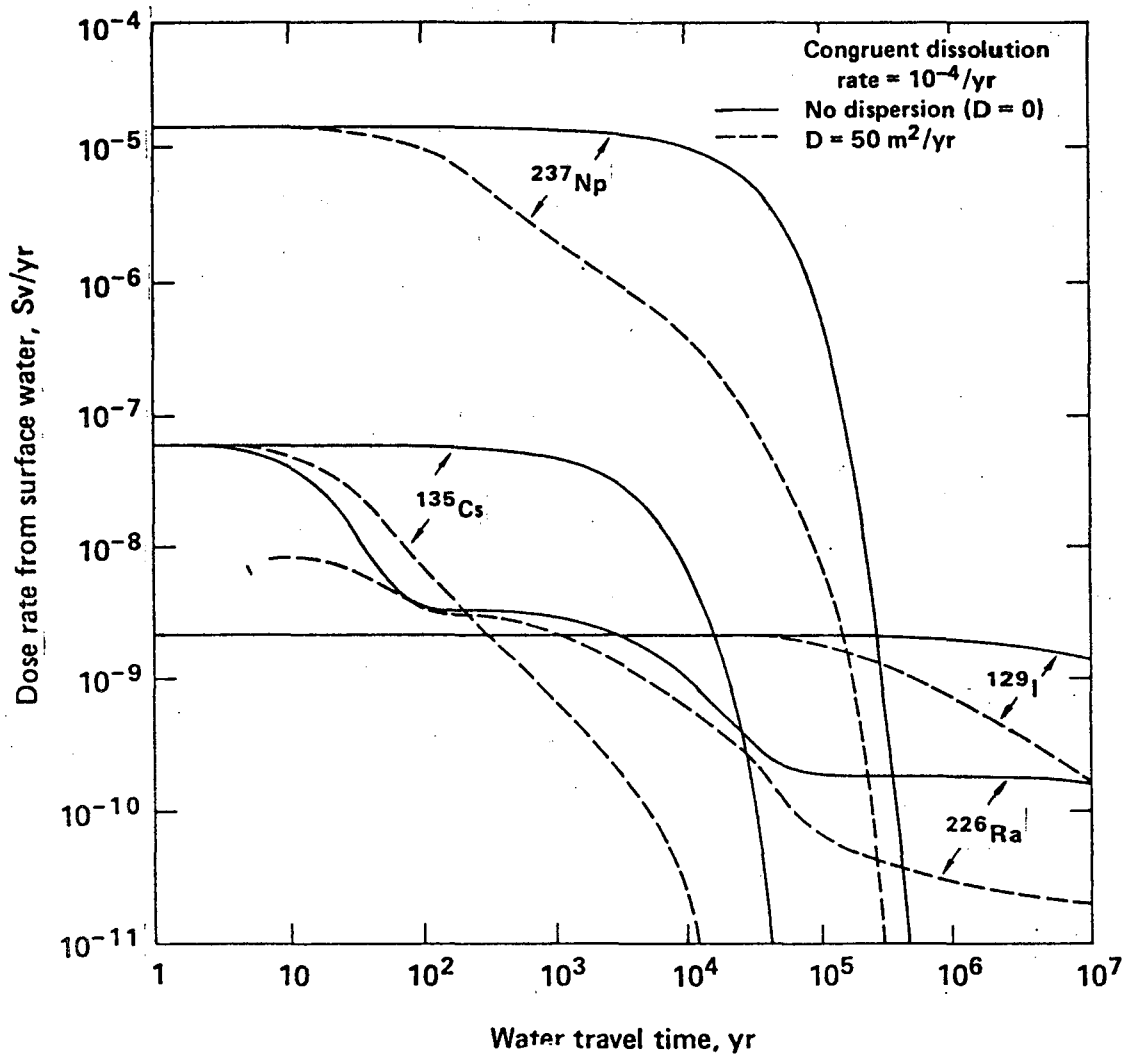
Figure 4. Individual radiation dose as a function of water
travel time for basalt, effect of dispersion,
reprocessing waste, congruent dissolution.
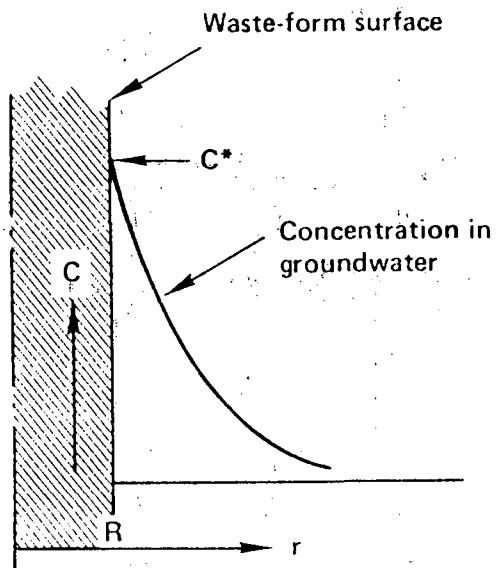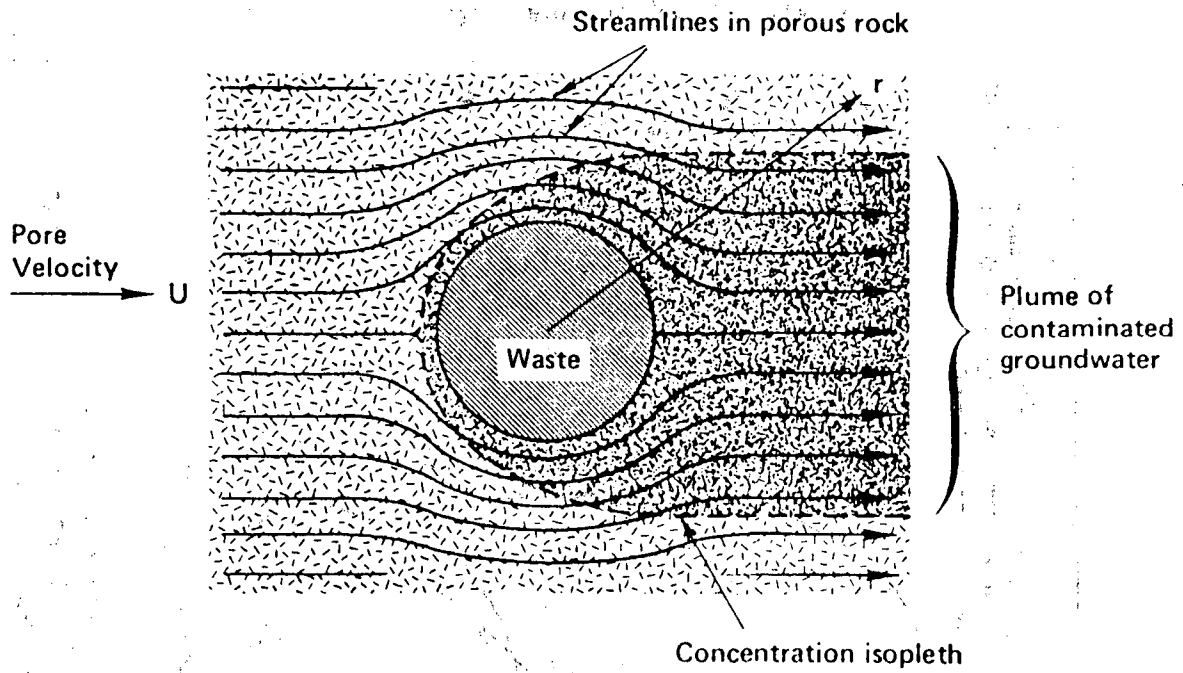
Figure 5. Velocity and concentration profiles for groundwater flowing across a waste cylinder.
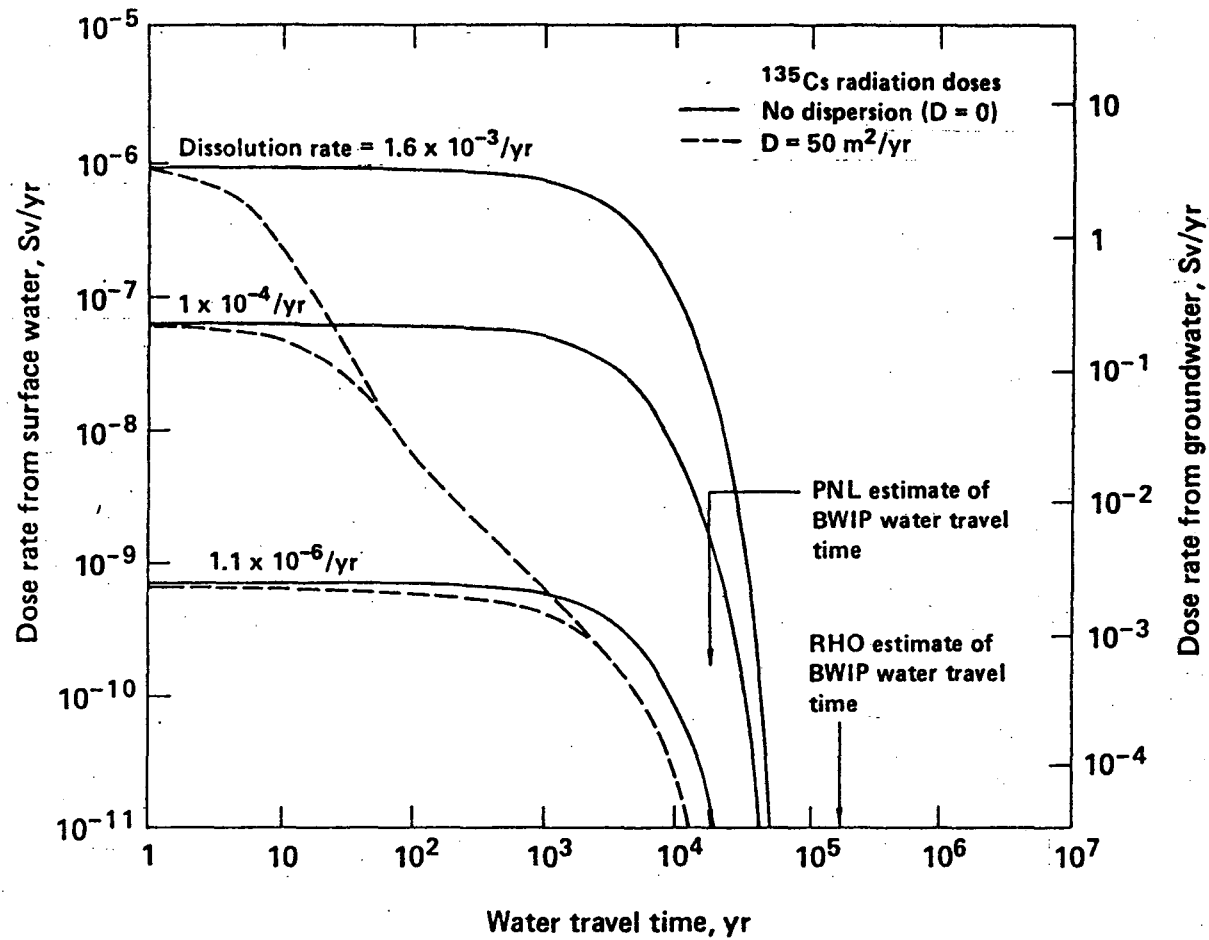
Figure 6. Individual dose from cesium-135 as a function of water travel time for basalt, effect of dissolution rate and dispersion, for reprocessing waste.
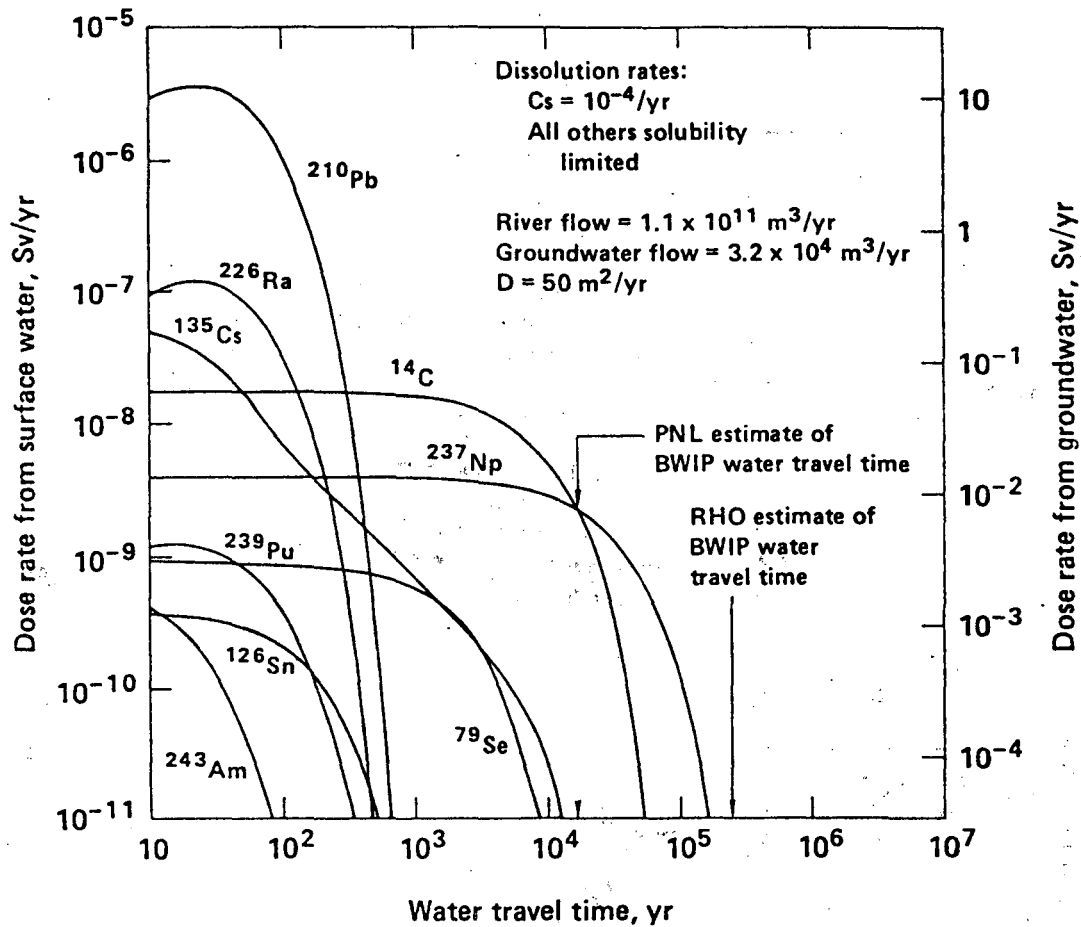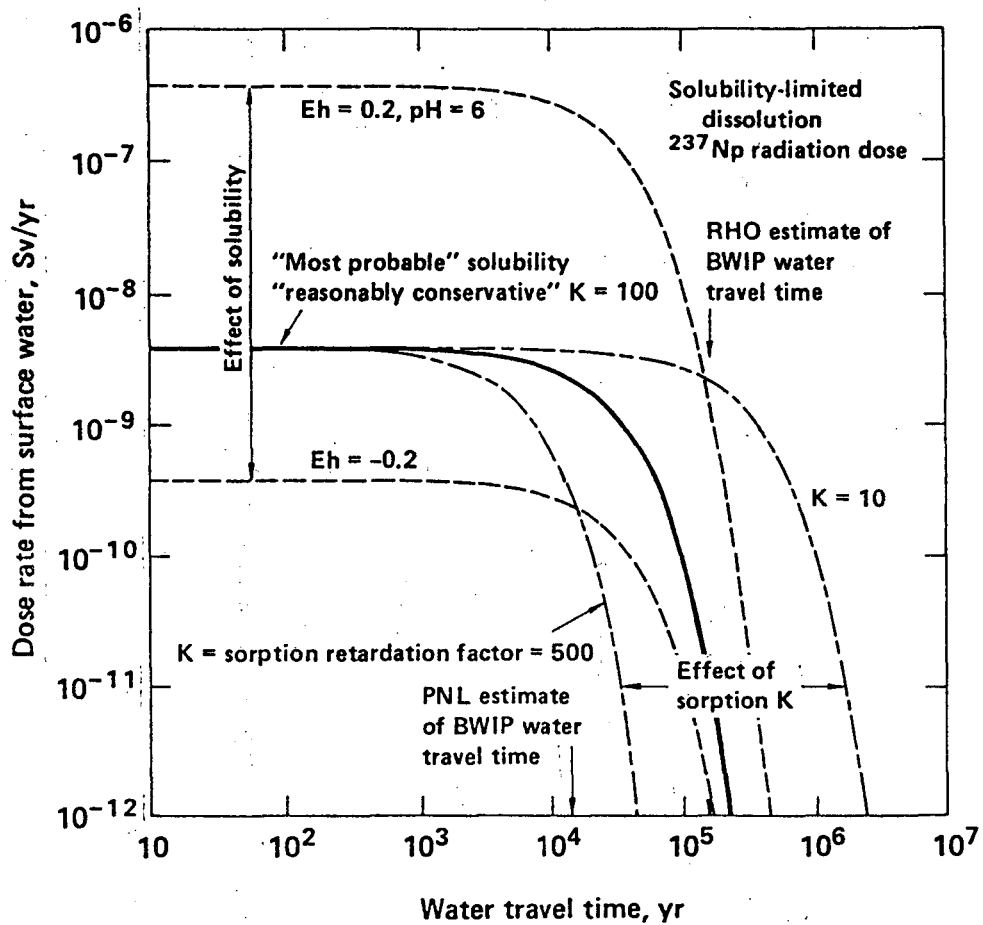
Figure 7. Individual radiation dose as a function of water travel time for basalt, reprocessing waste, solubility-limited dissolution.
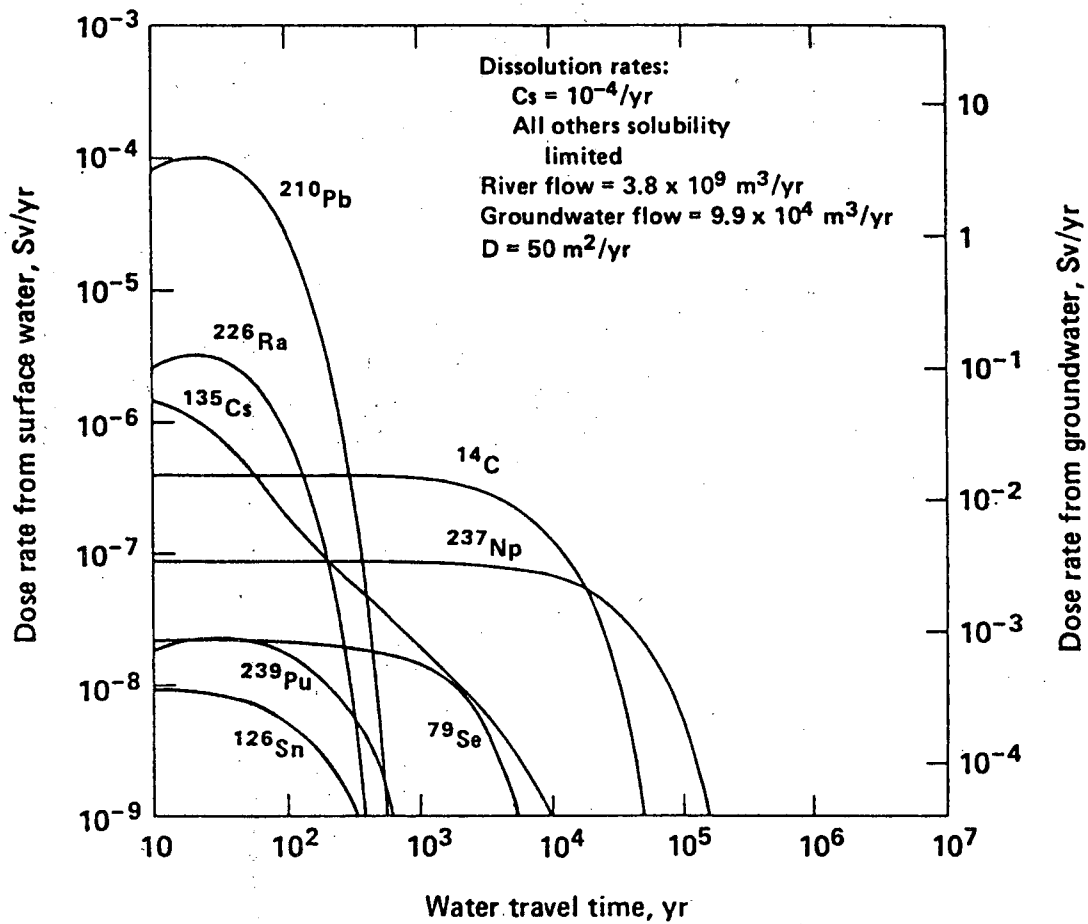
Figure 8.   Individual radiation dose as a function of
water travel time for granite, reprocessing
waste, solubility-limited dissolution.
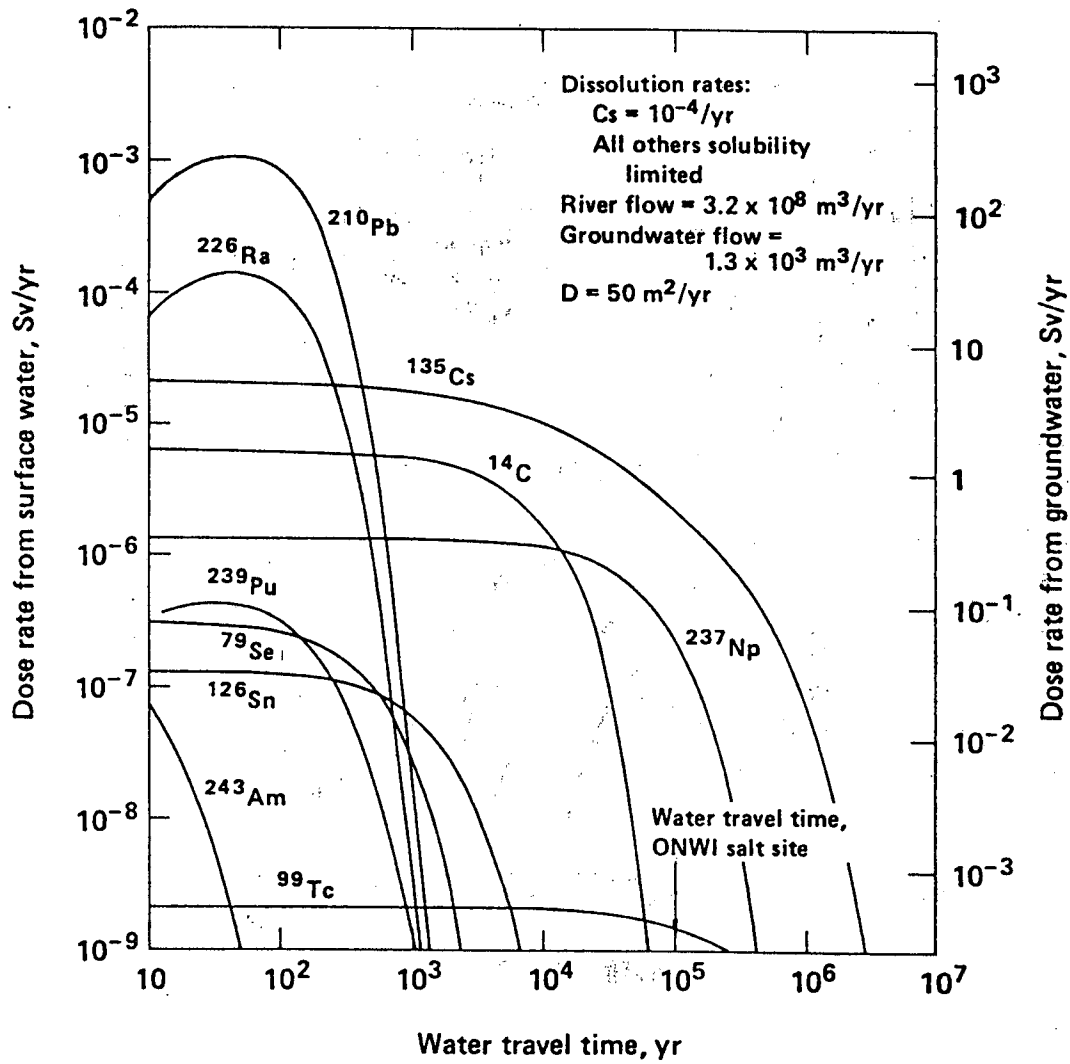
Figure 9. Individual radiation dose as a function
of water travel time for salt, reprocessing
waste, solubility-limited dissolution.
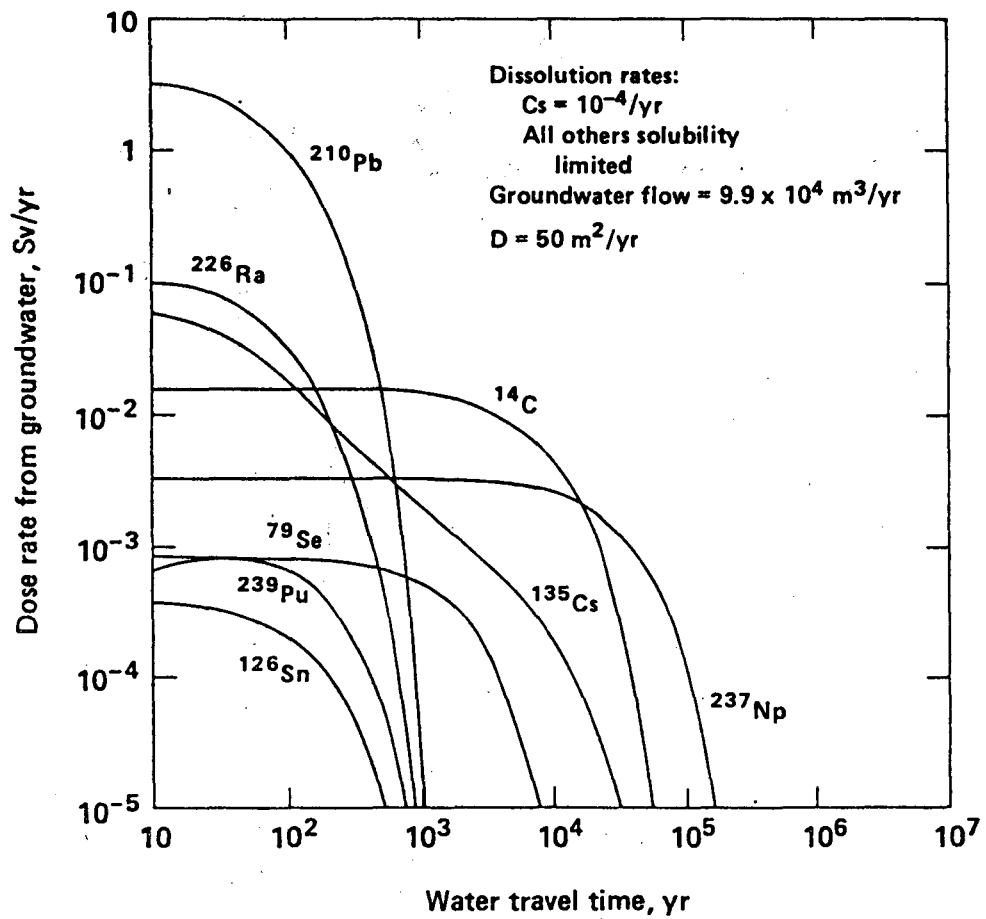
Figure 10. Individual radiation dose as a function of
water travel time for tuff, reprocessing
waste, solubility-limited dissolution.
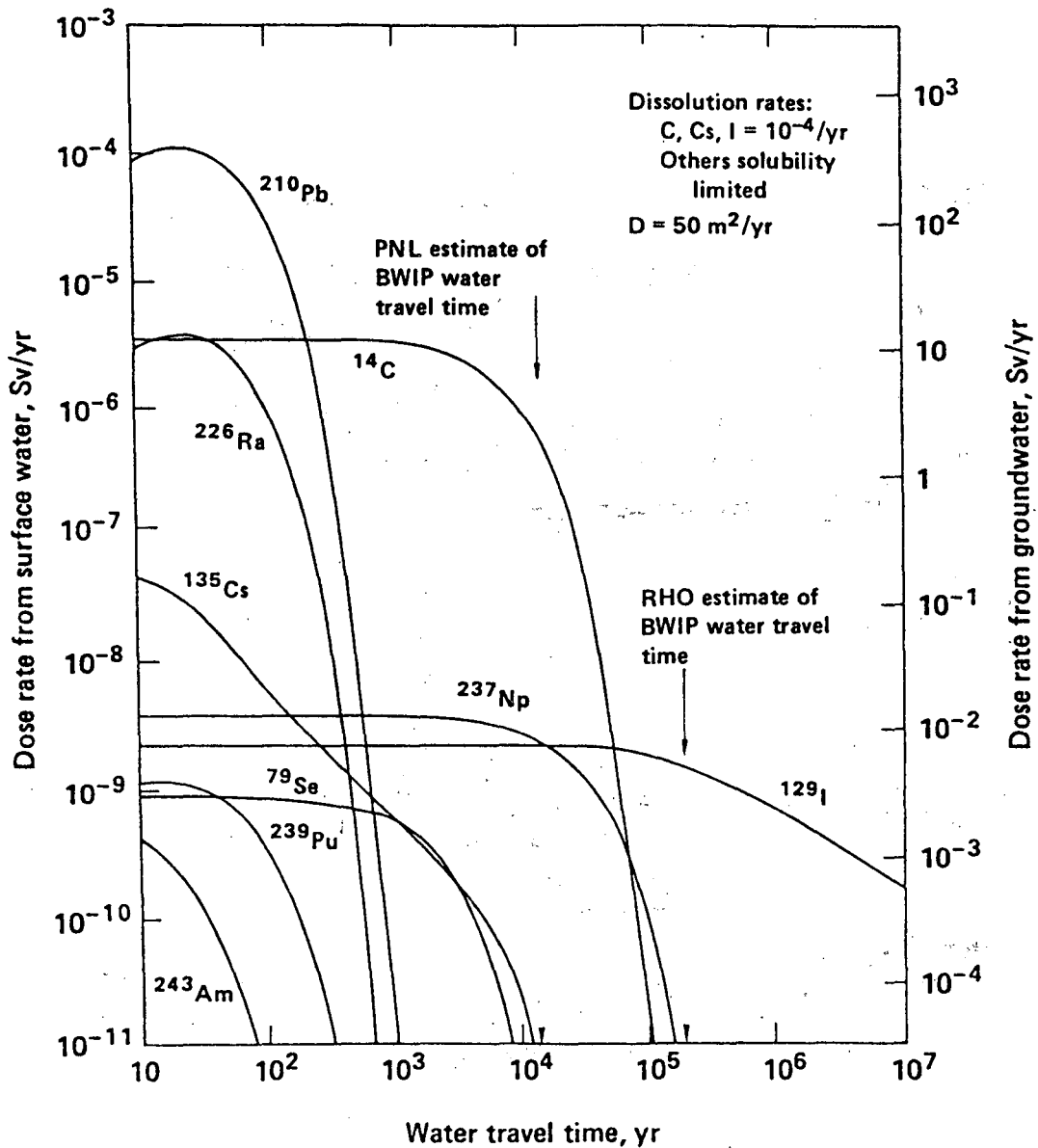
Figure 11. Individual radiation dose as a function
of water travel time for basalt, spent
fuel, solubility-limited dissolution.

Figure 12.   Individual radiation dose from neptunium-237
as a function of water travel time for basalt,
reprocessing waste, solubility-limited
dissolution, effect of uncertainties in
solubilities and retardation.