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DOSE-TO-MAN FROM SRP WASTE:
SENSITIVITY TO LEACHING AND ROCK PROPERTIES

INTRODUCTION AND SUMMARY

The risk of exposure to humans from high-level radioactive waste disposal may depend on the properties of the solid form containing the waste. An assessment is being made of the relative risks to man from the geologic disposal of alternative forms containing SRP high-level waste (HLW).

Perhaps the most complex features of the waste disposal system are the geologic repository and the overlying rock, which will act as isolation barriers to hinder the transport of radionuclides. It is generally assumed that the major risk of radionuclide release from a repository would be caused by leaching of the waste form, followed by the slow transport of waste elements by natural subsurface waters. This report summarizes results of a sensitivity analysis that predicts which features of the waste form and barrier system will have the greatest impact on dose to man.

Major conclusions of this sensitivity analysis are:

- Maximum dose never exceeds natural background radiation, and generally ranges from 10^{-4} to 1 mrem/yr.
- Waste forms with leach rates on the order of 10^{-5}/yr or greater will have no significant effect on dose.
- Leach rates significantly less than 10^{-5}/yr will decrease dose.
- Dose is insensitive to delays before leaching begins.
- Dose is insensitive to hydrologic dispersion.
- Actual doses could be much lower than those calculated if greater interaction occurs between radionuclides and the rock (e.g., a higher K_d).

DISCUSSION

Several studies have examined the transport of radionuclides to man after closure of a repository containing canisters of waste. These studies include transport models of varying sophistication, using incomplete data for chemical and physical properties of the waste and the rock. The purpose of SRL repository risk studies is to assess the relative performance of alternative waste forms under such conditions.

Test data are not available for alternative forms containing actual waste in repository environments. However, the effects of generic properties such as leach rate can be studied. Two major questions arise:

(1) Do lower leach rates result in proportionately lower doses?

(2) If dose can be decreased by using a more durable waste form, is this significant? Or, will this reduction lower an already insignificantly small dose?

The Hazard of SRP Waste

Potential radiologic doses from HLW would be much lower if releases were delayed until after certain radionuclides have disappeared. The relative radiological hazard* of the SRP waste inventory for times after closure of a repository is shown in Figure 1.[1]

*Hazard is calculated from Nuclear Regulatory Commission recommended conversion factors for critical-organ dose commitment after ingestion by an average adult.[2]

After about 300 years, ^{90}Sr and ^{137}Cs will have virtually disappeared, and the hazard will stabilize at values roughly three orders of magnitude lower than at the time of repository closure.

A peak occurs between 30,000 and 1,000,000 years, as ^{226}Ra accumulates from the decay of ^{234}U and ^{230}Th. Radium-226 is a significant hazard because it spawns eight short-lived daughter radionuclides.

Transport to Man

A detailed discussion of groundwater transport of waste species was given in a previous report.[3] In the release scenario, which is illustrated in Figure 2, some process causes groundwater to contact the waste form, dissolve it, and then transport waste elements to the earth's surface. The waste will interact with the rock during transport due to sorption, ion exchange, and precipitation. The major parameters which determine dose rates are:

Leach Initiation Time, tinit, is the time delay between repository closure and when the outer barriers fail and groundwater contacts the waste form. This period could correspond to the 1000-yr period of "zero release" suggested in NRC criteria.[1]*

Leach Duration, T, is the length of time needed to dissolve a waste form at a constant leach rate. The leach duration is the reciprocal of the fractional leach rate; a leach duration of 10^4 yr would satisfy an NRC criterion.[4]

Groundwater travel time, t_{gw}, is the time needed for water to travel from the repository to the biosphere.** The NRC has suggested 1000 years as a minimum t_{gw}.[4]

Retardation Factor, R, expresses how much interaction occurs between traveling waste elements and the rock. Each radionuclide will have its own retardation; the radionuclide travel time is equal to $R \times t_{gw}$. Conservative, baseline values of R for important elements in bedded salt are listed in Table 1.

Dispersivity, d, is a measure of how a release "pulse" is spread as waste is transported, by geologic dispersion and molecular diffusion. Dispersivity is measured in units of meters.

Dose Model describes what portion of the released waste is transported to man, how it is assimilated, and its biological effects.

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*In defining properties, a simple waste package design is assumed, as if a waste form in a long-lived canister were placed directly in the rock. "Leaching" refers to release of waste to the rock, which may be more complex than actual leaching because of backfill or getter materials present in the repository.

**The travel time depends on groundwater velocity and the length of the path; however, dose is more sensitive to the time of release than to small changes in path length and velocity,[3] so t_{gw} has been chosen as the variable.

A simple model has been chosen for biosphere transport and human uptake. Waste is released to a river of modest flow rate (10^9 m^3/yr). Human exposure is solely due to ingestion of 370 liters of water from this river per year. Ingestion of radionuclides is converted to dose by using the 50-year dose commitment factors to critical organs for a 1-yr exposure to an average adult.[2]

This model is used only to give a perspective of what range of doses can be expected; the same model was used by Koplik et al. to compare several published risk studies.[5]

RESULTS

Doses were calculated for various choices of parameters, using the equation* for fractional release:[5]

$$M = \left[T^2 + \frac{R^2}{V^2} \cdot 4\pi d \cdot L\right]^{-1/2} \quad \text{, where}$$

M = peak fractional release to the biosphere, yr^{-1}
V = groundwater velocity, m/yr
L = path length, m
d = dispersivity, m.

Values for V and L were chosen to span the range of observed groundwater velocities in geologic media. Assumed values for each choice of t_{gw} are listed in Table 2. Typical values of d range from 5 to 50 m.

The fractional release to the biosphere was converted to Curies/yr by multiplying by the Curie content of the source, corrected for radioactive decay during transport. The biosphere transport and dose model (above) was then applied to yield estimated dose-to-man.

Dose from a System Meeting 10CFR60 Criteria

A list of peak doses for a disposal system that just meets the minimum standards of the NRC's "Proposed Criteria for Radioactive Waste Repositories"[4] is given below. (Leach initiation time = 1000 yr; groundwater travel time = 1000 yr; and leach rate = 10^{-5}/yr.)

Dose (mrem/yr)	Time before biosphere release, yr	Radionuclide(s)
7.57×10^{-1}	59,500	^{226}Ra (from ^{234}U)
5.39×10^{-2}	59,500	Other U isotopes & daughters
3.70×10^{-4}	2,000	^{99}Tc
2.19×10^{-4}	232,000	^{237}Np
3.36×10^{-5}	462,000	^{93}Zr
1.16×10^{-5}	577,000	^{242}Pu, ^{239}Pu
2.35×10^{-6}	289,500	^{126}Sn

*This relation is an approximation of the solution to the one-dimensional transport equation for a single species, described in the earlier report.[3] The error due to the approximation was found to be undetectable when graphed. Radionuclide chains were assumed to travel through the geologic medium with the velocity of the longest-lived parent nuclide.

Effect of Groundwater Travel Time

A plot of dose vs groundwater travel time (t_{gw}) is shown in Figure 3. The packet of radionuclides transported as ^{234}U or ^{238}U (dominated by ^{226}Ra) generally provides the maximum dose for t_{gw} > 300 yr.

Effect of Leach Rate

Dose vs leach duration (T) is shown in Figure 4 for several choices of t_{gw}. Dose is insensitive to short leach durations, but becomes proportional to leach rate for extremely long T. Radium dose is not affected by leach rates worse than 10^{-5}/yr, while the dose from ^{99}Tc is always sensitive to leach rate.*

Doses for leach rates of 10^{-3}, 10^{-5}, and 10^{-7}/yr and several values of t_{gw} are shown in Figures 5 and 6 for radium-226 and technetium-99, respectively.

Retardation Factor

Release time (and thus dose) is greatly affected by the degree of retardation. Dose as a function of uranium retardation is plotted in Figure 7. The plotted range of R spans only the range of values that have been assumed for published risk studies.

Leach Initiation Time

As long as t_{gw} is greater than about 300 yr, dose is not sensitive to the time of leach initiation. Dose vs t_{init} is plotted in Figure 8.

Dispersivity

For all but very short leach durations, dose is not highly sensitive to dispersivity. This is illustrated for ^{226}Ra in Figure 9. Technetium is even less affected by dispersion, due to its small retardation.

Comparison with Previous Studies

Peak doses as a function of leach rate, using geologic properties from published studies and a 1000-yr delay before leaching begins, are shown in Figure 10 (salt) and Figure 11 (hardrock formations). Values of key parameters from these studies are listed in Table 3.

Even for extremely conservative geologic assumptions, predicted doses rarely exceed 1 mrem/yr.

*This effect is due to dispersion. If the radionuclide travel time is greater than the leach duration, enough time will elapse for release concentrations to be "dispersed". Tc is little retarded by rock; less time is available for dispersion to act.

FUTURE PROGRAM

This sensitivity analysis shows how dose would be affected by changes in geologic and waste form properties. Computer analyses using detailed models will yield more exact dose estimates. However, a full factorial analysis, varying all parameters across their credible ranges, would require a very large number of computer runs. Sensitivity analyses can fill gaps between selected calculations and thus extend the computer data.

ONWI has performed a preliminary analysis of dose from SRP waste in a salt repository.[6] That study used the PNL computer code GETOUT, which treats transport of full chains of radionuclides. Also included are detailed biosphere and food chain models.[7] The only parameters that were varied, however, were t_{gw} and t_{init}. The additional effects of leach rate, retardation, and dispersivity can be analyzed by using the sensitivity analysis.

LLL is conducting a comparative risk assessment of alternative waste forms for SRL. They will model waste form leaching in detail, and will vary most geologic parameters. Their code does not treat radionuclide chain transport, however, and suffers from insufficient data for geologic properties. Comparing the performance of different waste forms in geologic media other than salt can be accomplished by using a sensitivity analysis. A rough risk evaluation can be made before detailed geologic models are available.

These calculations can be used with waste inventories different from SRP feeds, to show whether one waste form or geologic medium would be attractive for a specific type of waste.

TABLE 1

RETARDATION FACTORS FOR BEDDED SALT

Values calculated from K_d values listed in Reference 9, using β of 11.5. K_d's are representative of salt, oxic conditions; retardation would be equal or higher for anoxic conditions. See discussion in Reference 3 (DPST-80-584).

Element	R	Element	R
Sr	12.5	Ra	69
Zr	461	Pa	576
Tc	1	U	58.5
Sn	288.5	Np	231
I	1	Pu	576
Cs	70	Am	1151

TABLE 2

VALUES ASSUMED FOR GROUNDWATER VELOCITY AND PATH LENGTH

Groundwater Travel Time t_{gw}	Path Length L	Groundwater Velocity V
100 years	707.1 meters	7.071 m/yr
300	1225	4.082
500	1581	3.162
1000	2236	2.236
3000	3873	1.291
5000	5000	1.0
10,000	7071	0.7071
30,000	12,250	0.4082
50,000	15,810	0.3162
100,000	22,360	0.2236
300,000	38,730	0.1291
500,000	50,000	0.1
1,000,000	70,710	0.0707

TABLE 3

GEOLOGIC PROPERTY DATA
USED IN PUBLISHED RISK STUDIES

	t_{gw}	G'water Velocity	Path Length	d	Retardation Factors Tc	U	Pu	Np
SALT MEDIA								
Cloninger (best properties)[9]	$3.8x10^5$	0.13	49,000	4.08	1	58.5	576	231
Logan[10]	$1.3x10^4$	1.5	20,000	50	1	21000	21000	160
WIPP EIS[11]	$5.6x10^3$	4.0	22,500	91	1	170	-	12000
ONWI baseline[6]	$1.0x10^4$	0.71*	7071*	4.08*	1	58.5	576	231
LLL median[8]	$1.0x10^4$	5.0	50,000	70	1	1000	1000	1000
HARDROCK MEDIA								
Berman[12]	$1.0x10^4$	1.6	16,000	50	1	10000	10000	10000
					Shale, Salt			
KBS[13]	$4.0x10^2$	5.75	2300	0.5	1	43	1100	260
					Granite			
Burkholder[14]	$1.5x10^2$	110	16,000	0.004	1	14000	10000	100
					Crystalline			
Hill & Grimwood[15]	$9.1x10^1$	110	10,000	30	1	14000	10000	100
					Crystalline			

*Assumed, by referencing earlier uses of that computer code (Ref. 9).

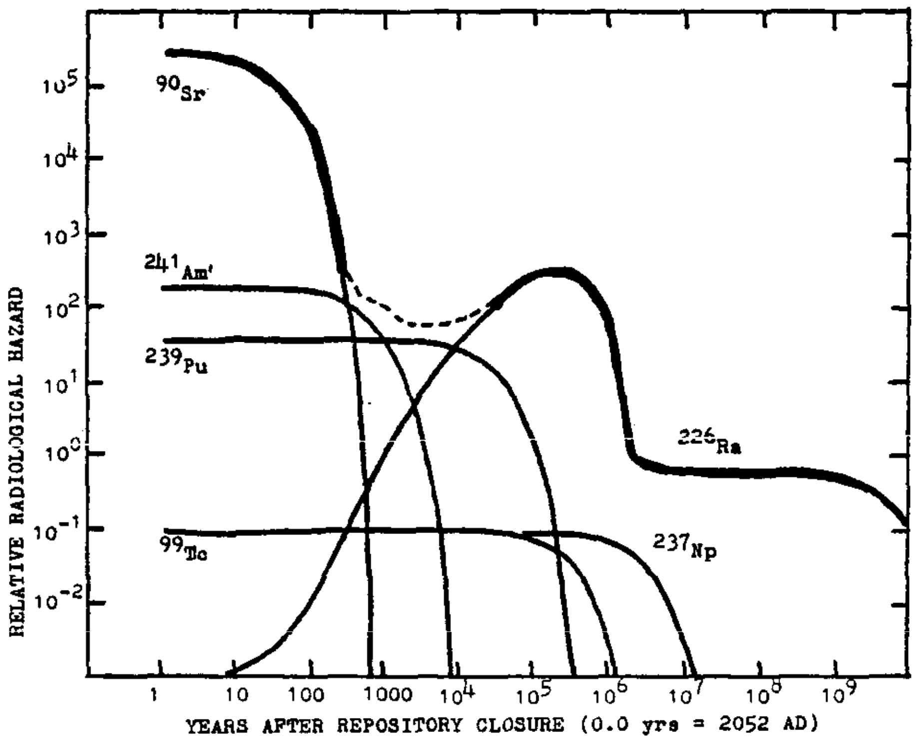

Figure 1. Radiological Hazard From SRP HLW

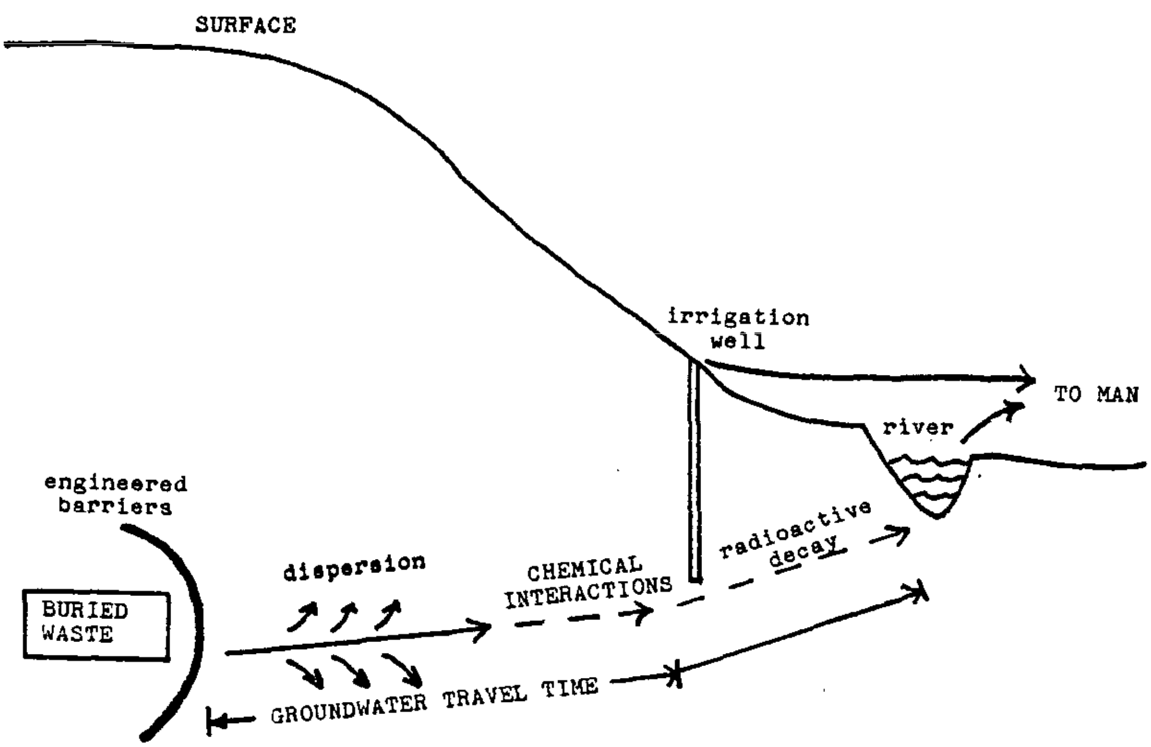

Figure 2. Schematic of Groundwater Transport

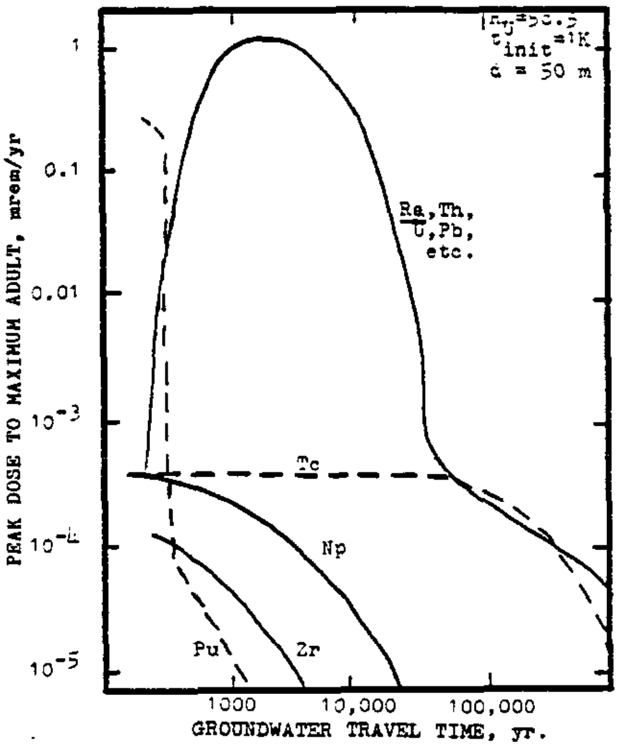

Figure 3. Effect of Groundwater Travel Time (t_{gw})

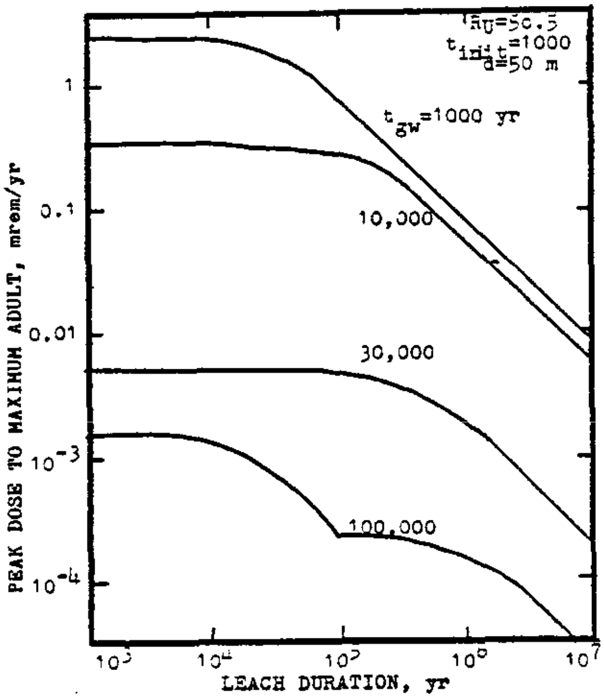

Figure 4. Effect of Leach Duration

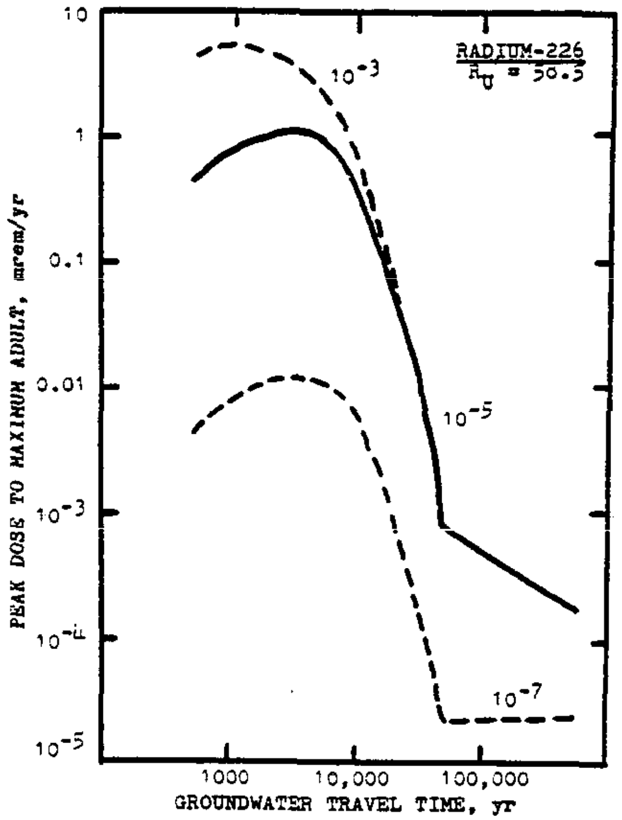

Figure 5. Effects of Groundwater Travel Time and Leach Rates for Radium-226

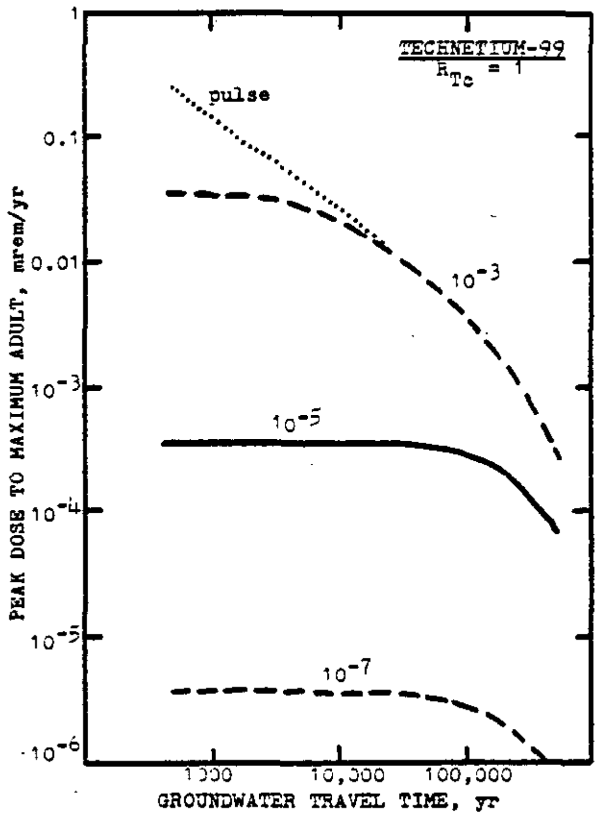

Figure 6. Effects of Groundwater Travel Time and Leach Rates for Technetium-99

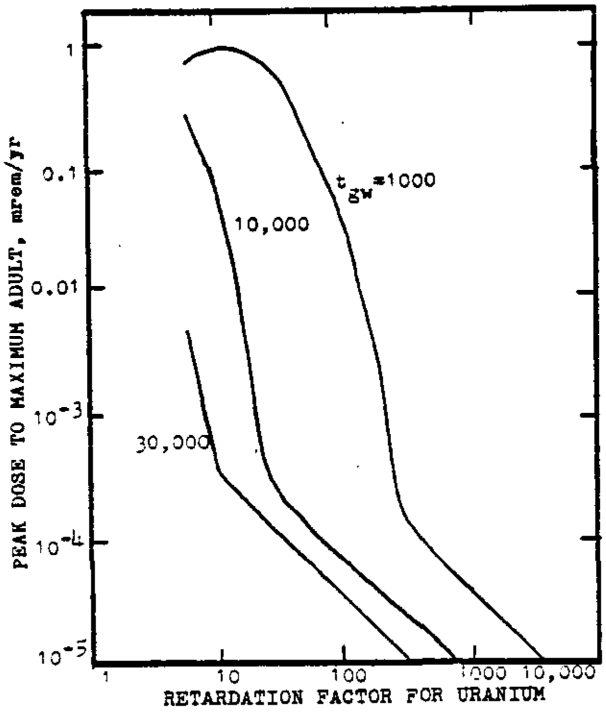

Figure 7. Effect of Retardation Factor (R)

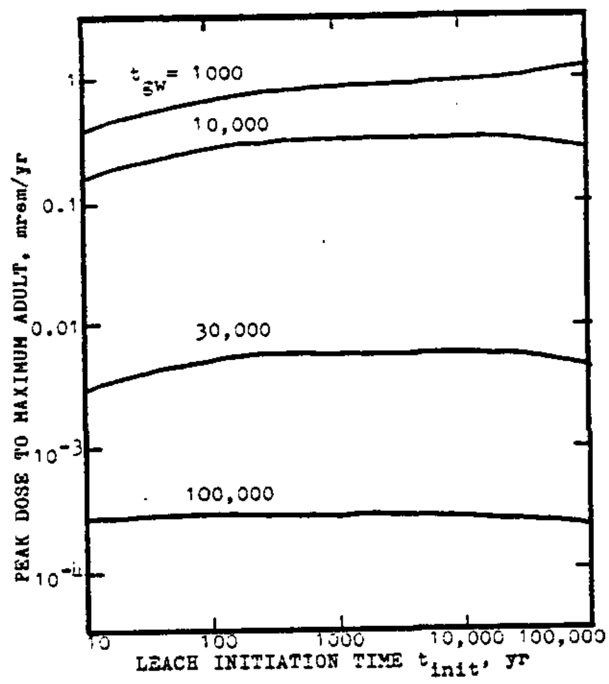

Figure 8. Effect of Leach Initiation Time (t_{init})

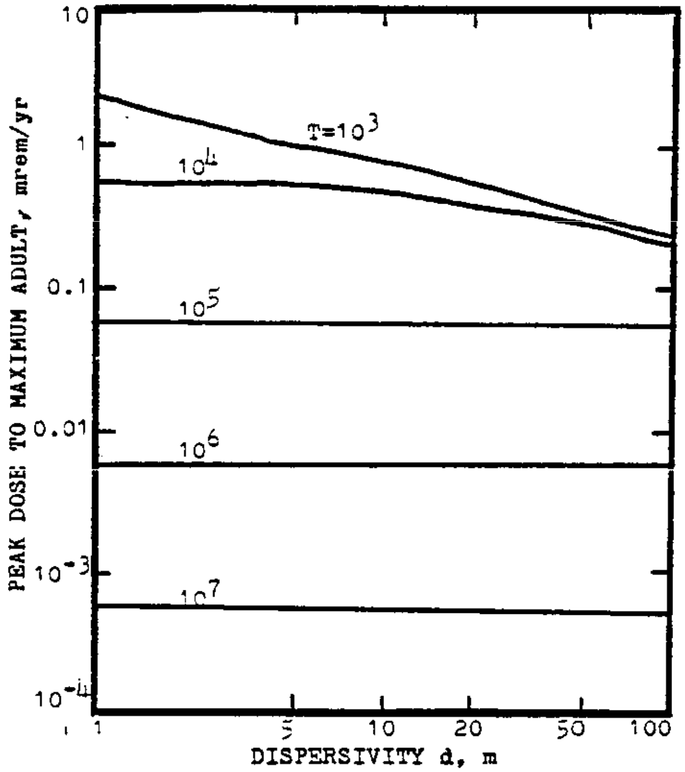

Figure 9. Effect of Dispersivity (d)

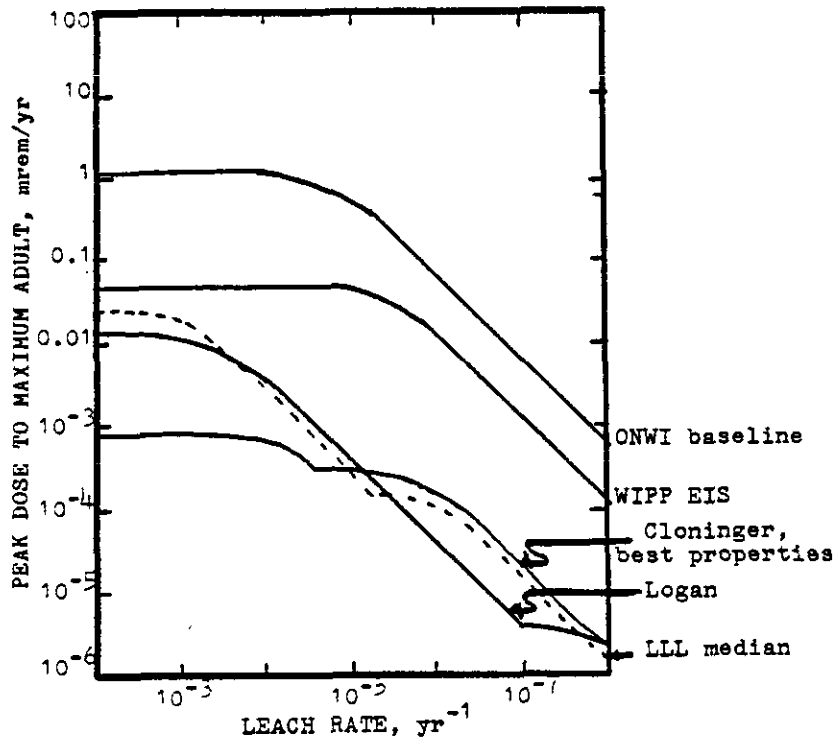

Figure 10. Doses Predicted for Properties of Salt Used in Published Risk Studies

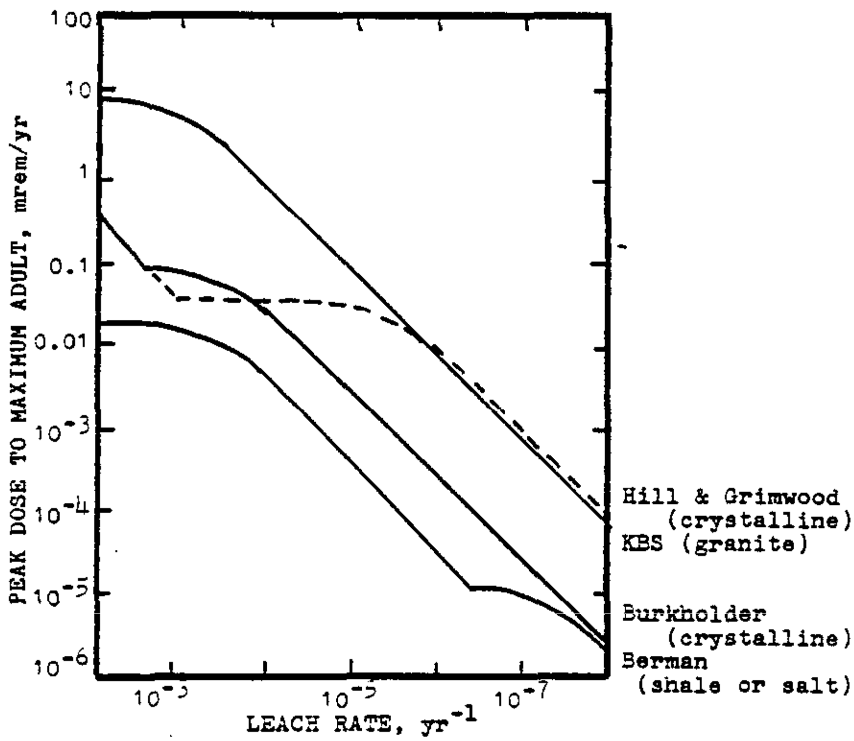

Figure 11. Doses Predicted for Properties of Hardrocks Used in Published Risk Studies

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