COMPARISON OF POTENTIAL RADIOLOGICAL CONSEQUENCES FROM A SPENT FUEL REPOSITORY VERSUS NATURAL URANIUM DEPOSITS[a]

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INTRODUCTION

It has been suggested that one general criterion for deep geologic repositories is that they should result in no greater radiological risk than that due to naturally occurring uranium deposits. This analysis[1] has been designed to investigate the reasonableness of that suggestion and to determine whether current expectations of spent fuel repository performance would be consistent with such a criterion. Reference spent fuel repositories were compared to natural uranium ore deposits both in terms of their intrinsic characteristics such as radionuclide inventory, depth, proximity to aquifers, regional distribution, etc., and in terms of radiological consequences that are now occurring from some ore deposits and may eventually occur from the repositories and from other ore deposits.

INTRINSIC PROPERTIES COMPARISON

The earth's environment is naturally radioactive due to its original inventory of elements including uranium and its daughters. Natural uranium deposits are analogous in many respects to the proposed spent fuel repository. There are also significant differences between the two systems.

Inventory and Concentration

The geologic disposal of radioactive waste, in the long term, does not change the natural inventory of uranium and its daughters, disregarding the fraction fissioned in a nuclear reactor. In the short term, the short-lived radioactive fission

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(a) Work sponsored by the Office of Nuclear Waste Isolation under its contract with the US Department of Energy.

products add significantly to the radioactive inventory. However, uranium and its daughter radium-226 (226Ra, and its daughters) are the toxic elements of major long-term concern and share this property with natural deposits. Also of long-term concern for the repository are the long-lived fission products 99Tc and 129I, and the actinide 237Np and its precursor 241Am, which, for all practical purposes, do not exist in the natural underground deposits, and 14C, which can occur in ore deposits in shallow sedimentary strata.

The inventory of 238U in the reference repository[2] is 24,000 Ci or 72,000 tonne of uranium. The quantity of uranium is dispersed over 8 km^2, 600 m below the surface in the form of 156,000 spent PWR fuel elements with a density of approximately 40 m^2 per 460-kg fuel element. The zone occupied by the fuel elements is 3.8 m thick; if the uranium were uniformly distributed throughout the zone, its average concentration would be about 0.16%.

The principal natural radioactive isotopes found in the earth's crust are 238U, 235U, 232Th, 40K, and 87Rb plus radioactive daughter isotopes such as 230Th, 226Ra, 222Rn and 231Pa. Other natural radioisotopes result from cosmic-ray bombardment of the earth's atmosphere, producing 3H (tritium), 14C and others. All radioactive elements spontaneously decay to stable elements in a series of stages. Once equilibrium is established the radioactivity associated with every stage is equal. Thus the decay chain of 238U consists of 14 stages, and the total radioactivity at equilibrium is 14 times that associated with 238U decay to 234Th. The decay chains of the other principal isotopes behave similarly, although only the 235U and 232Th have several stages in their decay series.

The concentration of radioactive elements in the earth's crust is subject to wide variation depending upon the rock type, basic rock chemistry, and the rock's geographic location. According to a survey of over 2000 analyses of rocks from around the world, by H. A. Wollenberg of Lawrence Berkeley Laboratory, University of California, the concentration of uranium in rocks ranges up to 720 ppm and of thorium up to 880 ppm.

The total uranium inventory of the earth is estimated as 6.3×10^{13} tonne or 2×10^{13} Ci. This is equivalent to the uranium inventory in 880 million reference repositories. The uranium

inventory of the United States is estimated as 2.4×10^{12} tonne and is equivalent to that in 34 million repositories. If currently mineable-grade uranium is 300 ppm or greater, the cumulative amount of uranium in the United States is 19 million tonne and is equivalent to the uranium content of 260 repositories[3]. January 1, 1978 DOE estimates of potential uranium resources available at $50 per pound of U_3O_8 is 2.7 million tonne uranium[4] or 0.9 million Ci, which is equivalent to that in 37 reference repositories.

The economic uranium occurs in concentrations of ore pods or roll fronts often along the interface between an oxidized and reduced area of sandstone. Uranium concentrations in the ore range from a lower economic mining limit of perhaps 0.05% up to as much as 14% U_3O_8. Individual ore pods may vary from an average of 3000 tonne of ore containing perhaps 5 tonne of uranium to single, large ore bodies of up to 10 million tonne of ore containing 1000 tonne of uranium. Thus, the mineable concentrations of uranium ore deposits are as large as or larger than the quantity of uranium dispersed in the storage zone of a nuclear-waste repository. This information is summarized in Figure 1. A single, small ore pod containing 6 tonne of uranium is equivalent to the uranium in 12 fuel elements and may have a projected area of 400 m^2 versus 490 m^2 for 12 fuel elements, however, the uranium concentration in natural deposits never approaches that within a fuel element.

While the repositories will contain significant inventories of other nuclides (which are treated in greater detail in the radiological consequences comparison later in this paper), the uranium series is a primary source of potential radiological dose and accounts for a major fraction of the radioisotope inventory in both the natural ore deposits and, especially for the long-term, the repositories.

Distribution and Proximity to the Surface

Uranium content of the whole earth is estimated to be about 11 ppb. However, most of this uranium has been concentrated in the mantle and crust by geologic processes.

Volcanic tuffs and breccias are important sources of uranium. These acidic rocks containing from 5 to 300 ppm uranium have been ejected into the atmosphere from volcanos and deposited within layers over thousands of square miles.[5] Granitic rocks

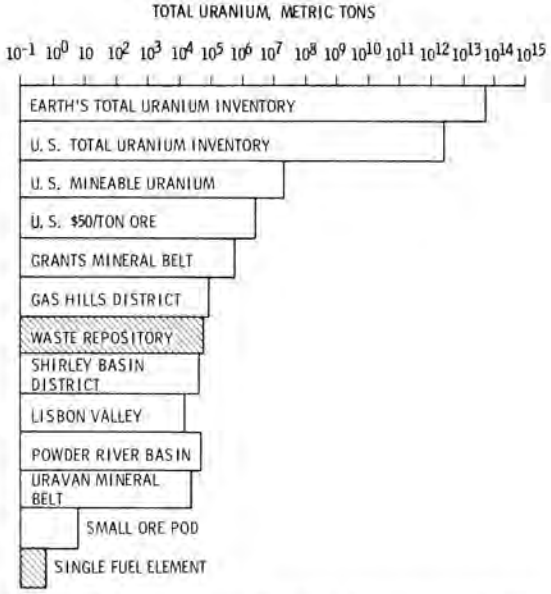

Fig. 1. Comparative Uranium Inventory of Waste Repository with Inventory of Naturally Occurring Uranium Deposits

exposed over large areas and containing from 5 to 30 ppm uranium are the source of many uranium deposits near the rock outcrops. Some of the granitic rocks are regarded as ultimate resources of uranium and thorium that could be exploited at tolerable costs. Uranium in the oceans is precipitated in organic muds or phosphate sediments. Marine shales are exposed or underlie large areas in the central and eastern United States. Approximately 4600 uranium deposits in sandstone host rocks outcrop or underlie huge areas of the west central states of New Mexico, Colorado, Utah, and Wyoming and are the major source of U.S. mined uranium and uranium reserves.

Uranium deposits can occur at the surface or at any distance below the surface. Uranium is being recovered from depths in excess of 3,000 m as a by-product of gold mining in South Africa. The uranium deposits at Oklo, Gabon are thought to have been buried as deep as 30 km by sediments before being exposed near surface by uplift and erosion. Knowledge of uranium at great depth is limited

Table I. 1977 U.S. Uranium Production: Distribution by Depth of Ore[4]

Depth of Ore, m	Uranium, tonne	Percent of Total	No. of Sources
0 - 30	690	5	105
30 - 60	3,200	25	60
60 - 90	1,600	13	355
90 - 120	1,700	13	20
120 - 150	770	6	20
150 - 180	1,100	8	7
180 - 210	840	7	15
210 - 240	310	2	4
240 - 800	2,400	19	11
Others	310	2	37
Total	13,000	100	314

because exploration is not economical. The distribution of uranium production in 1977 by depth of ore is shown in Table I. While 19% of the uranium mined in the U.S. in 1977 came from depths comparable to that proposed for spent fuel repositories, 79% of it came from rich deposits at depths significantly closer to the surface than is currently planned for the repositories.

Presence of Water

The primary long-term hazard from either a spent-fuel repository or a large ore deposit is due to the possibility that ground water may transport radioactive elements to the surface. The presence of water, its quantity, flow rate, and chemical composition are factors that govern the extent of the hazard. Water is present in rock at considerable depths occupying pores and fractures. Sedimentary rocks contain water at depths of 15 km and hard igneous rocks at perhaps 5 km. Depths greater than 30 km are at temperatures and pressures that close all pores and openings, and water is present only in chemical combination with rock minerals. Depths at which entry of water is precluded are too great for practical consideration.

The formation and migration of uranium ore deposits is shown pictorially in Figure 2. Oxygenated ground water enters at the recharge area and dissolves uranium. Dissolved uranium is then

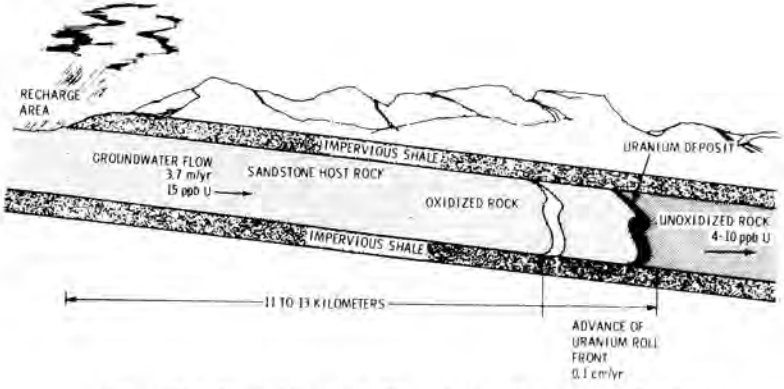

Fig. 2. Formation and Migration of Uranium Deposits in Sandstone Host Rocks

reduced by carbonaceous matter and sulfides; the reduced form precipitates, forming a deposit. Eventually, uranium is redissolved at the trailing edge of the roll front and transported to the advancing edge where it again precipitates. The advance rate of a roll front, measured by Russian geologists with similar rates accepted by American geologists, is on the order of 0.1 cm/yr. At this rate, roll fronts in Wyoming, which are found 11 to 13 km from the nearest recharge area, have been migrating for 11 to 13 million years. Thus, the deposit has been in a state of "failure" for several million years.

The concentrations of uranium and radium in ground water in the United States range up to 800 ppb with a median of about 1.5 ppb for uranium and up to 29 pCi/L with a median close to 0.3 pCi/L for radium. Surface waters (rivers/lakes) have an average uranium content of 0.1 ppb and the oceans about 1 ppb.[3]

A significant amount of natural radioactivity exists in ground and surface waters and, in fact, the richest deposits of uranium and thorium occur in past and presently active aquifers. While a repository for spent fuel will, by design, be located in dry strata away from aquifers, there is a finite probability that at some future time ground water may enter the repository and begin to transport radionuclides to the surface. However, as the preceding discussion indicates, the presence of radionuclides at

significant concentrations in our ground and surface waters will be nothing new. The following section on potential radiological consequences comparison addresses this subject more explicitly.

RADIOLOGICAL CONSEQUENCES COMPARISON

This second comparison is based on the potential long-term radiological consequences that may occur from the repositories versus the existing and potential radiological consequences from natural ore deposits. The basis for this part of the analysis was methodology developed[6] for estimating the potential radiological consequences due to a scenario involving ground water intrusion, leaching and nuclide transport. The scenario has two subsets, a general scenario and an intrusion water well scenario. The general scenario consists of release to surface water (lake or river) as the biosphere entry point. The human-caused intrusion water well scenario was chosen as a limiting case of the ground water pathway. This methodology was applied in a limited manner to predict the potential radioactive release from large uranium deposits and further to compare these predictive calculations with earlier calculations[7,8] for spent fuel repositories on an equivalent basis. The consequences are expressed in terms of average annual, total-body dose to a "maximum individual."

Approach and Methodology

To develop a representative scenario for naturally occurring uranium ore deposits, two uranium deposits were selected to serve as references. The two are Granite Mountain in Wyoming and Rossing in Namibia. The Granite Mountain deposit is a widely distributed, low concentration deposit, while the Rossing deposit is a more compact, higher concentration deposit.

The characteristics of the Granite Mountain deposit are assumed to be those of 60 million years ago. Some granite samples from the Granite Mountain have lost nearly 80% of the uranium originally contained in them.[9] Assuming that all the leached uranium was returned to the deposit, the uranium content would be about 10^{11} kg. It was assumed that the 238U daughters are in decay equilibrium.

The Rossing deposit in Namibia (formerly South West Africa) produces nearly 4000 tonne/yr uranium from 44,400 tonne/day of ore containing 300 ppm uranium. The total uranium content in the

deposit is about 6 x 10^7 kg. The deposit is in the Namib Desert near the edge of the Khan River Basin, but the river contains water only when there is runoff from the infrequent rains.

The inventories of dose-significant nuclides for the two uranium deposits and the reference repositories are listed in Table II. A summary of the descriptive data for the geologic transport calculations is in Table III. The descriptive data for the uranium ore deposits were combined with other data needed for the nuclide-transport calculations. The methodology was applied using a new FORTRAN version of the GETOUT[10] nuclide-transport code. The biosphere-transport/dose code, PABLM[11], is the same as that used in CWM/GEIS[2]. These models were applied to predict the potential radiological consequences from the two example ore deposits in a manner consistent with the analyses of the reference repositories.[7,8]

Table II. Radionuclide Inventories

Nuclide	Initial Inventories, Ci[a]		
	Reference Repositories[b]	Granite Mountain	Rossing Deposit
^{238}U	2.4+4	3.4+7	1.9+4
^{234}U	1.3+5[c]	3.4+7	1.9+4
^{230}Th	2.9+1	3.4+7	1.9+4
^{226}Ra	2.9-1	3.4+7	1.9+4
^{129}I	2.5+3	--	--
^{99}Tc	9.6+5	--	--
^{14}C	2.5+4	--	--
^{241}Am	2.3+8	--	--
^{237}Np	2.5+4	--	--

(a) 2.4+4 is notation for 2.4×10^4, etc.
(b) from CWM/GEIS[2], with 1/5 of the total commercial no-recycle inventory in one repository
(c) Includes ingrowth from 238Pu during first 100 years.

Table III. System Parameters for Geologic Transport Calculations

Site	Ground Water Velocity, m/yr	Migration Pathlength km	Release Rate, Fraction/yr
Granite Repository	1.5-2 to 2.0+0	1.0+0 to 1.5+1	1.0-3 to 1.0-6
Salt Repository	1.7-1 to 1.1+1	5.6+0 to 6.4+1	1.0-3 to 1.0-6
Granite Mtn	2.2-1 to 4.5-1	2.3-1 to 3.2+1	1.3-7 to 1.3-8
Rossing	1.1+0	5.5-1 to 3.2+0	1.0-7

Uranium and radium are found in almost every source of surface or subsurface water. Selected uranium and radium concentrations found in ground and surface waters of the United States were used in conjunction with the biosphere model to estimate the potential dose consequences due to a maximum individual's use of these waters for all water needs. This dose was based on either the reported 226Ra concentration or on the assumption that 226Ra is in decay equilibrium with the reported uranium concentration. The uranium is assumed to be totally 238U. It has been found that normal ground water generally contains a higher specific activity (Ci/L) of radium than uranium[12] partly due to reconcentration[13], enhanced solubility due to α-recoil and solution chemistry; thus the use of the equilibrium assumption is conservative.

Results

The results are summarized in Fig. 3 and 4, which show a comparison among calculated dose from the measured radionuclide concentrations in U.S. waters, the calculated potential releases from the two ore deposits, and potential releases from the two reference repositories. Fig. 3 shows the results for the general scenario. Fig. 4 shows the results for the intrusion water well scenario. The total body dose from the ore deposits and for most of the parameter range of the repositories is dominated by 226Ra created from uranium parents. Included in this dose is the direct

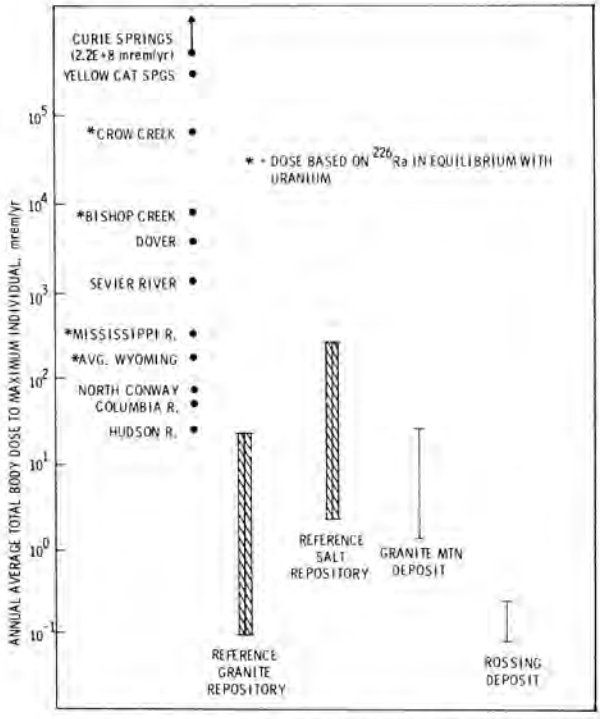

Fig. 3. Summary of Radiological Consequences: Average Annual Total-Body Dose, General Scenario

dose from 226Ra and its daughters. The uranium itself usually contributes a minor fraction of the total dose. However, under some conditions for the reference granite repository, 234U plus plus 237Np from its original inventory and from its precursor, 241Am, contribute significantly to the total body dose. For very extreme release rates of nuclides into the ground water, 14C, 99Tc and 129I can dominate the dose from both repositories.

With the exception of the upper bounds of potential general scenario consequences from the reference salt repository (Fig. 3), the results from the repositories are in the same general range as those from the ore deposits. Note that most of the points representing the measured surface water concentrations lie above

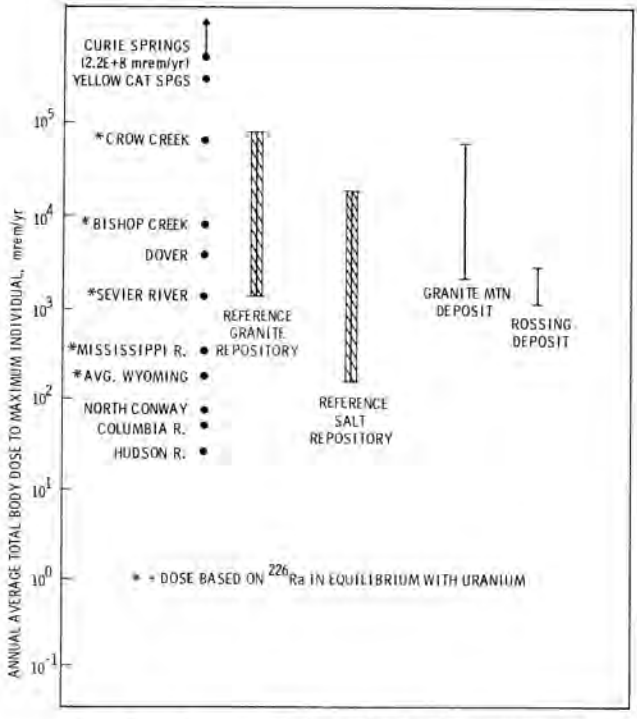

Fig 4. Summary of Radiological Consequences: Average Annual Total-Body Dose, Well Scenario

the range calculated for both reference repositories and both ore deposits. Two reasons for this are apparent (other than the uncertainties regarding scenarios and the geologic transport calculations). First, these measured values represent resultant consequences from quite a range of actual yet unknown scenarios in terms of release rate from the ore, size and quality of the ore deposit, ground-water velocity, and proximity of the deposit to the discharge point where the measurement was made. Second, many of the measured values probably represent waters whose radionuclide content is not from a single source, as were the calculations for the repositories and ore deposits, but from

several in addition to the minerals from the immediate area where the samples were taken.

As might be expected, the dose consequences from the intrusion water well scenario, summarized in Fig. 4, are considerably higher for both repositories and both ore deposits, and they generally lie in the higher range of the measured values. Furthermore, the reference repositories and the ore deposits are even more similar in potential consequences from the well scenario than from the general scenario summarized in Fig. 3. The extremely high well scenario doses for the repositories, ore deposits, and the conversions of nuclide concentrations in U.S. waters that are reported here are based on linear extrapolation from dose transfer factors that were determined at much lower dose rates. Thus, the actual upper limits for the well scenario consequences from the reference repositories and the ore deposits as well as those for extremely high reported nuclide concentrations (Crow Creek, Yellow Cat Springs, and Curie Springs) would, in reality, be somewhat lower; however, relative to one another, they would still be about the same.

CONCLUSIONS

On the basis of the intrinsic properties comparison, the long-term hazard from spent fuel in a geologic repository is comparable to that of a large ore deposit, although the specific nuclide concentration in an individual fuel element is not duplicated in nature.

On the basis of the second comparison, a repository constructed within reasonable constraints presents no greater hazard than a large ore deposit. Even without such constraints, however, the natural hazard due to some observed radioactive releases to the biosphere in the United States far exceeds any that could reasonably be expected from a spent-fuel repository.

While at first intuition it is reasonable to use natural ore deposits as a basis for criteria for nuclear waste repositories, the variability in the natural system is so great that it does not allow a specific criterion to be stated in absolute terms. It is suggested that if the naturally radioactive environment is to be used as a basis for a criterion regarding repositories, the criterion should be based on the radiological quality of the environment in the immediate region of a specific repository, and

should be in terms of an acceptable potential increase in that radiological content due to the existence of the repository. This criterion could then be used as a basis for developing, through experimentation and modeling, specific criteria for a repository system design that is reasonably assured of meeting the environmental criteria.

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