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AN ASSESSMENT OF ISSUES RELATED TO DETERMINATION OF TIME PERIODS REQUIRED FOR ISOLATION OF HIGH LEVEL WASTE

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ABSTRACT

A commonly held perception is that disposal of spent nuclear fuel or high-level waste presents a risk of unprecedented duration. In 40 CFR 191, the EPA requires that projected releases of radioactivity be limited for 10,000 years after disposal with the intent that risks from the disposal repository be no greater than those from the uranium ore deposit from which the nuclear fuel was originally extracted. This study reviews issues involved in assessing compliance with the requirement. The determination of compliance is assumption dependent primarily due to uncertainties in dosimetric data, and relative availability of the radioactivity for environmental transport and eventual assimilation by humans. A conclusion of this study is that, in time, a spent fuel disposal repository such as the projected Yucca Mountain Project Facility will become less hazardous that the original ore deposit. Only the time it takes to do so is in question. Depending upon the assumptions selected, this time period could range from a few centuries to hundreds of thousands of years considering only the inherent radiotoxicities. However, if it can be assumed that the spent fuel radioactivity emplaced in a waste repository is less than 1/10 as available for human assimilation than that in a uranium ore deposit, then even under the most pessimistic set of assumptions, the EPA criteria can be considered to be complied with.

INTRODUCTION

Because of the extended half-lives of many of its components the disposal of high-level nuclear waste (HLW), and spent nuclear fuel (SNF) is commonly believed to present a problem of unprecedented duration (1, 2). In previous studies (3, 4) it was found that this belief does not have a sound logical or technical basis since:

[1] There is an inverse physical relationship between half-life and specific radioactivity such that the longer the half-life of any radionuclide, the less radioactive it is, and

[2] The stable toxic elements (e.g., Pb, Cd, etc.) do not elicit a comparable degree of concern regarding their safe disposal despite the fact that they will remain toxic forever (5).

Nevertheless the concept that HLW requires long-term isolation is well embedded in the public's mind, reflected in regulations, and therefore, must be addressed in any sound management plan.

Several previous studies (6, 7, 8) have explored the relative hazards of HLW to determine suitable periods for its isolation. The most prominent approach involves comparison of the relative hazard of the waste against that of the uranium ore from which nuclear fuel had originally been extracted.

The question of determining required isolation periods has again been raised in regard to the projected emplacement of spent nuclear fuel at the Yucca Mountain Project Facility. This study is intended to address that question.

BACKGROUND

Long-term isolation requirements for the disposal of HLW in a deep geologic repository were developed by the Environmental Protection Agency (EPA) (9). These criteria establish that projected releases of radioactivity be limited for a period of 10,000 years. The EPA release criteria were based on the assumption that if the repository could provide adequate containment of the material for 10,000 years, it is reasonable to assume that it would provide adequate containment for periods beyond this time.

For the geologic repository, the two primary release pathways are [1] the long-term movement of nuclear material to the repository boundary and beyond by groundwater; and [2] human intrusion into the repository horizon. In effect, the second pathway is very scenario-specific and the risk is of very low probability. Therefore, this study will focus on the groundwater release pathway.

In 40 CFR 191, EPA states that radioactivity release from the waste disposal facility should be such that "risks to future generations will be no greater than the risks that would have existed if the uranium ore used to create the wastes had not been mined to begin with." To make such a determination requires a comparative assessment of the SNF or HLW repository against a typical uranium ore deposit. The objective of this study is to make such an assessment using the most recent data on radionuclide inventories and dose consequences.

COMPARATIVE ASSESSMENT

To compare the potential hazard of the repository against that of the uranium ore deposit from which the nuclear fuel was originally extracted requires the determination of a sound basis for comparison. Some possibilities include comparative evaluation of hazard per unit mass (repository vs. ore body) or per unit volume. Such evaluation, however, would involve use of relatively subjective assumptions regarding dimensions and other properties. A more objective basis would involve comparison per metric ton of heavy metal (MTHM) considering the uranium ore quantities and components required to produce 1.0 MTMH of nuclear fuel against those in the corresponding quantity of spent fuel. That is the basis for comparison applied in this study.

Radioactivity in Uranium Ore

Determination of the quantity of ore required to produce 1.0 MTHM must consider all losses in processing the ore into nuclear fuel. For purposes of this evaluation, it is assumed that: 90% of the U_3O_8 is recovered from the ore, 1.0% is lost in conversion of U_3O_8 to UF_6, the uranium is enriched from natural uranium to 3.2% U-235 with a 1.0% material loss in the enrichment process, and other small losses in the processes of conversion to and fabrication of nuclear fuel. Table I summarizes the radionuclide content of the uranium ore required to produce 1.0 MTHM of nuclear fuel. For purposes

of comparison, it is conservatively assumed that the radionuclides in uranium ore are its only hazardous components. Stable toxic elements such as lead are ignored.

Table I

Radioactivity in Spent Fuel

The radionuclide content in spent fuel was determined using the ORIGEN-2 code (10) calculations. For purposes of comparison it is assumed that the fuel was fissioned in a PWR to a thermal burnup of 33,000 MWD. The radioactivity at four selected post-irradiation times is shown in Table II. Only those radionuclides constituting at least 1.0% of total radioactivity at any given time are listed.

Table II

Method of Comparison

Various methods for comparison of the relative hazard of waste repositories to uranium ore deposits can and have been applied. These methods fall into two basic categories: [1] calculational models which predict maximum radiation doses to individuals or populations, and [2] relative hazard indices. Calculational models have been applied by Wick and Cloninger (11) and by Williams (12). Both of these studies indicate the hazard from the HLW repository is comparable to or less than that of typical uranium ore deposits.

Hazard Indices have previously been applied for the purpose in several studies including: Smith et. al. (13), Haug (14), Tonnessen & Cohen (15), Klingsberg & Duguid (16) and McGrath (17). These studies also indicate that, in time, the relative hazard of the HLW or SNF repository becomes less than that of the uranium ore deposit. The time periods required for this condition to occur range from a few centuries to several millennia depending upon the approach and assumptions applied.

In this study, the hazard index approach is applied including more recent data on spent fuel composition from the ORIGEN-2 code and updated dosimetry data from the International Commission on Radiological Protection (ICRP). The objective is to evaluate and provide perspective on the time required for the hazard potential of the spent fuel repository to decrease to that of a uranium ore deposit.

HAZARD INDEX CALCULATIONS

Several different hazard indices for evaluation of the relative hazard of waste disposal operations have been proposed and/or applied. These range from simple indices which consider only the inherent toxicity to complex indices which also consider the potential for the toxic materials to migrate and be assimilated by humans (i.e., availability), persistence of the hazardous material, and other factors which might affect the overall hazard. In a comparative assessment such as this study, as a first approximation, it

is reasonable to assume equivalent availability between the entities being compared (i.e. repository vs. ore body) and focus on relative toxicities. Relative availabilities can then be evaluated as a subsequent step. Therefore, a simple hazard index is used consisting of the quotient of the quantity of material divided by its allowable exposure limit according to appropriate standards/criteria for allowable exposure. In this study the relative hazard index (RHI) of spent fuel is evaluated against that of an equivalent quantity of uranium ore such that:

$$RHI = \frac{\sum \left(\frac{(Q_i)}{(EL_i)} \right) \text{(Spent fuel)}}{\sum \left(\frac{(Q_i)}{(EL_i)} \right) \text{(Uranium ore)}}$$

where:

RHI = Relative Hazard Index
Q_i = Quantity of Radionuclide i
EL_i = Allowable Exposure Limit for Radionuclide i

Where RHI>1.0, the spent fuel has an inherent radiotoxicity exceeding that of the corresponding uranium ore.

Exposure Mode

The consequences of exposure to radioactive materials are dependant upon the exposure mode. For example, the dose consequences of ingested can be quite different than from inhaled radioactivity. For radioactivity buried underground as is the case with both the repository and the ore deposit, the predominant exposure mode is clearly via ingestion. Accordingly, only hazard resulting from the ingestion mode is considered.

Exposure Limits

As new information and insights are obtained regarding dosimetric effects of radionuclides, allowable exposure limits have been revised. Current standards in the USA as specified in 10 CFR 20 reflect the recommendations of the International Commission on Radiological Protection (ICRP) as published in their ICRP-2 report in 1959 (18). The recommended ICRP-2 exposure limits are prescribed in terms of a maximum permissible concentrations (MPC's) for individual radionuclides.

In 1980, the ICRP issued its revised recommendations for exposure limits to radionuclides in its ICRP-30 report (19) incorporating more recent information on dosimetric effects. These recommendations prescribe an allowable limit of intake (ALI) for ingested radionuclides. In previous studies, hazard indices based on ICRP-30 indicate a significantly longer time period for repositories to reach the equivalent hazard of a typical uranium ore body (6) (7) (8). This was largely the result of an apparent increase in predicted dose consequences from Np-237 and somewhat reduced consequences

from Ra-226. Although predicted dose consequences from certain other radionuclides were also revised, the changes in Np-237 and Ra-226 had the greatest effect. These changes were based upon new data indicating a new understanding of the fraction of ingested radionuclide assimilated as described by the f_1 factor. For example, ICRP-30 assumed a 100 fold increase for fractional assimilation (f_1) of Np-237 over that assumed in ICRP-2.

Later, ICRP again revised f_i, factors for certain radionuclides in their ICRP-48 report (20) issued in 1986. These later changes were reflected in the proposed revision of 10 CFR 20 and in draft DOE Order 5480.XX (21). In the latter document, exposure limits are prescribed in terms of derived concentration guides (DCG's).

Calculation

Using the data and approach described in the previous sections, a calculational model was developed to determine relative hazard indices for radionuclide components of spent fuel vs. uranium ore as a function of time. This model utilized a spreadsheet program and was developed to run on an IBM-PC computer. To allow for comparison, calculations were performed based upon application of ICRP-2, ICRP-30, and ICRP-48 recommendations.

CALCULATIONAL RESULTS

The calculational results are presented in Fig. 1. Based upon dosimetry data derived from ICRP-2, the apparent crossover time (time required for the spent fuel to become less hazardous than uranium ore) is about 500 years. This result is consistent with that determined in previous studies (14) (15) (16). ICRP-30 dosimetry indicates that although the initial hazard level is somewhat less than from ICRP-2, the crossover time is extended to over 100,000 years. Results based upon ICRP-48 dosimetry are essentially similar to those based upon ICRP-30.

FIGURE 1

From Fig. 1 it can be seen that the dosimetric data assumed in the analysis can have a significant effect upon the apparent crossover time. However, in all cases, the relative hazard of spent fuel will, eventually, become less than that of the corresponding uranium ore.

DISCUSSION

Other information that can provide insight on the relative hazard of SNF vs. Uranium ore includes comparative evaluations on the toxicities of natural uranium and its fission products, relative availabilities including the effect of engineered barriers, and a consideration of the geotoxicity hazard index.

Natural Uranium and Its Fission Products

To gain further perspective on the crossover phenomenon a calculation was performed considering only pure natural uranium. In this calculation, 1 Kg of natural uranium (0.7% U-235) is evaluated for an extended period of 10^{15} years under two conditions. These conditions are: complete fissioning

vs. natural decay of all uranium atoms. The results are shown in Fig. 2. The solid line shows the hazard index of the products resulting from fission of every atom in 1 Kg of natural uranium including the stable end products of decay (Sn, Ba, Zr, etc.). The solid line shows the hazard index for the products of natural decay reflecting increased toxicity due to ingrowth of radium and eventually the formation of stable lead. For this calculation, ICRP-2 dosimetry is used, and for stable elements, EPA drinking water standards are used to indicate relative toxicity. It can be seen that even in this extreme case (pure uranium) a crossover occurs at about one million years. Given enough time, it is apparent that the fissioning of uranium will eventually result in a net decrease the earth's toxic burden.

FIGURE 2

Availability

Previous discussion has considered only inherent toxicities of materials assuming ingestion. The spent fuel vs. uranium ore comparisons presume that spent fuel in a waste repository with all of its natural and engineered barriers would, nonetheless be equally as available for environmental transport and eventual assimilation by humans as would the natural uranium distributed more or less randomly throughout the earth's crusts by nature (neither NEPA nor NWPA were in effect at the time of Creation).

If we make the reasonable assumption that spent fuel as emplaced in a reasonably well-designed repository would be somewhat less available to the biosphere than would naturally occurring uranium, the net effect would be to displace the curves in Fig. 1 proportionate to the degree of diminished availability. Figure 3 presents this concept using the most recently determined (and most conservative) dosimetry (ICRP-48).

FIGURE 3

If, for example, spent fuel in a repository were 1/10 as available as naturally occurring uranium ore (i.e., relative availability = 0.1) the crossover time would be ~10,000 years. With a relative availability of 0.01 the crossover time would be ~1,000 years, and with a relative availability of 0.001, the relative hazard of the spent fuel in a repository would be less than that of uranium ore at all times. To provide some perspective on relative availabilities, the effects of natural and engineered barriers as proposed for the repository design should be considered.

Barriers

The repository differs from a natural ore body in that as part of the siting and design process, a number of engineered and natural barriers will be utilized as mechanisms for mitigating the movement of the radioactivity to the accessible environment as depicted in Fig. 4. The goal of the siting process is

FIGURE 4

to select a location for the repository which will provide substantially better containment than a natural ore body; in effect, a site that specifically meets or exceeds the requirements promulgated by 40 CFR 191. At the direction of Congress, the potential site at Yucca Mountain in southern Nevada is being characterized to determine if it can meet the regulatory criteria. Accordingly, this discussion will focus on barriers specifically applicable to the projected Yucca Mountain repository.

The first barrier is the waste itself. After the first 2,000 years, the waste material is typically in the form of an oxide or metal of very insoluble transuranic elements. Since the movement of material would be through groundwater, it can be reasonably assumed that the availability of the radioactive material to the accessible environment would be reduced by at least a factor of 10.

The second barrier is the waste package. During the first 1,000 years (the period in which the fission product component of the waste decays away), the waste package is projected to provide essentially complete containment. Containment during this period is particularly important since most of the fission products in the waste are generally much more soluble than the longer-lived transuranic radionuclides. In effect, from a waste isolation standpoint, the primary function of the waste package is to remain intact for 1,000 yrs and contain the more soluble fission products.

The third barrier is the location of the repository relative to the groundwater system. The repository site is selected so that there will be no groundwater flow through the underground waste storage horizon. Since groundwater is the predominant transport mechanism for the nuclear material, no release will occur unless water is present. Despite the emphasis on careful site selection however, it is possible that 1) there exists a potential for a geologic change to alter groundwater flow paths, or 2) our presumed knowledge regarding the site characteristics may be in error. However, at the Yucca Mountain site, these possibilities are extremely remote based on current knowledge. Continuing site characterization efforts will likely provide further assurance that the possibility for water intrusion is essentialy zero.

The fourth barrier is also related to repository site selection. Criteria considered for site selection include attributes which would: 1) minimize inadvertent human intrusion into the repository, and 2) limit the potential for geological activities which may bring the material to the surface. Because of these siting considerations, it is projected that the potential for release of radioactivity to the accessible environment due to such events should be reduced by at least two orders of magnitude relative to that from a uranium ore deposit.

The fifth barrier is the characteristics of the local groundwater system. Groundwater in the Yucca Mountain area is typically very limited in volume and basic in pH. Limited volume means that very little waste material will be dissolved because there is minimal water for areal saturation and

dissolution of the waste. The potential for radionuclide migration via groundwater transport is further reduced by the fact that the pH of the water is high, decreasing the solubility potential of the material in the water. In addition, the geologic media through which the groundwater moves will tend to reduce the transport of solutes by various physical-chemical processes including ion-exchange and adsorption. This will not only decrease the quantity of material that could leach into the groundwater, but will also prolong its potential travel time beyond the repository boundary.

The sixth barrier is the groundwater travel time. Since groundwater movement is extremely slow in the Yucca Mountain region, it will take between 10,000 and 100,000 years for the material to move beyond the repository boundary. It is estimated that this attribute coupled with the characteristics of the groundwater system should allow for an overall time of at least 100,000 years to reach the accessible environment.

The seventh barrier includes institutional controls such as markers and records, which document the location of the repository. This barrier will help minimize the potential for inadvertent human intrusion.

Geotoxicity of Materials

In a previous study (22) a complex hazard index was developed to describe the relative hazard of toxic materials buried underground by either man or nature. This Geotoxicity Hazard Index (GHI) considers the inherent toxicity, availability, and persistence (half-life) of materials. Figure 5 presents the GHI for various materials assuming equivalent burial conditions (10 meter burial depth, no engineered barriers). The relative hazard potential of these relatively familiar materials can provide general perspective. A recent study by Brookins et al. (23) can provide a more specific insight. This study indicates that surface soils in the vicinity of the Yucca Mountain site at certain playa areas (dry lakes) could have significantly greater inherent toxicity than uranium ore. This condition is due to high concentrations of toxic minerals such as arsenic, selenium, vanadium, and boron. At certain times, standing surface waters in such areas can approach lethal concentrations of toxic materials (so called "poisoned water holes"). Further study on these Nevada playas may provide greater insights on comparative potential risks to health from natural and man-made entities in the Yucca Mountain region.

FIGURE 5

SUMMARY AND CONCLUSIONS

EPA requirements for disposal of spent nuclear fuel, as specified in 40 CFR 191, limit projected releases of radioactivity to the "accessible environment" for a period of 10,000 years after disposal. In 40 CFR 191, this requirement is described as "a time frame without precedent environmental regulations." The need for long-term isolation of HLW and spent nuclear fuel reflects the common belief that these materials present a potential hazard to the environment of unprecedented duration. Although this belief may have no logical basis, it is nonetheless so deeply embedded that it must be addressed in any plan for HLW management.

This study has explored certain issues related to determination of time periods required to assure isolation of HLW, and addressed various approaches toward making such a determination. Of these approaches, the one most commonly accepted and endorsed by the EPA is to assure that risks to future generations are no greater than the risks that would have prevailed if the uranium ore used to create the nuclear fuel had not been extracted from the earth to begin with.

The comparison of relative risks from spent nuclear fuel in a repository to that of an equivalent quantity of uranium ore is assumption dependent. In evaluating the relative risks, the most important assumptions that must be made are: [1] The dosimetric basis for evaluating the relative radiotoxicities of the radionuclide mixtures in spent fuel vs. that in uranium ore, and [2] assessment of the relative availability for environmental transport and assimilation by humans. In this study, the relative risks have been evaluated using a variety of assumptions. From this evaluation, it is concluded that regardless of the assumptions used, the risk to public health from a HLW or spent fuel waste repository will always become less than that of the original uranium ore deposit. Only the time it takes to do so is in question. Useful insights may also be derived from comparative assessments against certain native Nevada soils and mineral deposits which could pose significantly greater risks than would a uranium ore body.

The crossover time (i.e., time required for the repository to become less hazardous than the uranium ore deposit) can vary from a few centuries to hundreds of thousands of years, depending upon the assumptions used. Using the most conservative dosimetric data (ICRP-48), if it can be assumed that the radioactive contents of the repository are less than 1/10 as available for release to the biosphere as those in a uranium ore deposit, the EPA requirement for limiting projected radioactive release for a 10,000 year period can reasonably be assumed to be met. Considering the nature of the many barriers to release that are included in the repository design, this should easily be the case.

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TABLE I

URANIUM ORE REQUIRED TO PRODUCE 1.0 MTHM OF NUCLEAR FUEL*

RADIONUCLIDE	Ci	MASS (Kg)
Pb-210	2.3	--
Bi-210	2.3	--
Ra-223	0.1	--
Ra-226	2.3	--
Th-227	0.1	--
Th-230	2.3	--
Th-239	2.3	--
Pa-231	0.1	--
Pa-234	2.3	--
U-234	2.3	--
U-235	0.1	50
U-238	2.3	6,900

* ASSUMING 0.2% URANIUM ORE, TOTAL MASS OF ORE REQUIRED $\approx$ 35,000 TONS

Table II

SIGNIFICANT RADIONUCLIDES* IN SPENT NUCLEAR FUEL

(PWR, 33,000 MWD/MTHM Burnup)

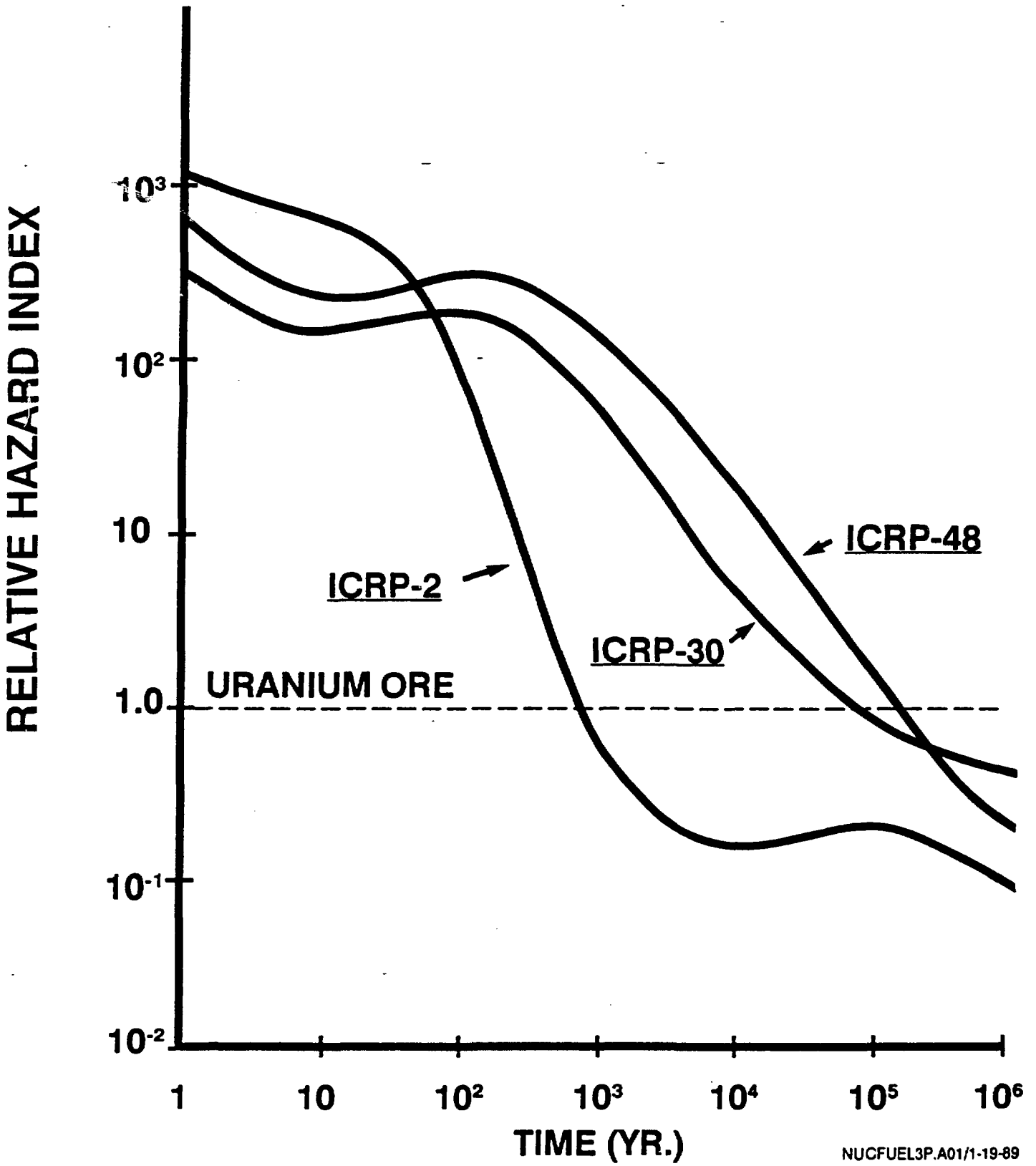
Radionuclide	Ci/MTHM			
	1 yr	100 yr	10^4 yr	10^6 yr
Ni-59			4.7E0	
Sr-90	7.1E4	6.7E3		
Y-90	7.1E4	6.7E3		
Zr-93				1.2E0
Zr-95	3.1E4			
Nb-93M				1.2E0
Nb-95	7.1E4			
Nc-99			1.2E1	5.0E-1
Ru-106	2.7E5			
Cs-134	1.1E5			
Cs-135				2.6E-1
Cs-137	1.0E5	1.0E4		
Ba-137	9.6E4	9.7E3		
Ce-144	4.5E5			
Pr-144	4.5E5			
Pm-147	1.0E5			
Pb-209				9.1E-1
Pb-210				4.6E-1
Pb-214				4.6E-1
Bi-210				4.6E-1
Bi-213				9.1E-1
Po-210				4.6E-1
Po-213				8.9E-1
Po-214				4.6E-1

Radionuclide	Ci/MTHM			
	1 yr	100 yr	10^4 yr	10^6 yr
Po-218				4.6E-1
At-217				9.1E-1
Rn-222				4.6E-1
Fr-221				9.1E-1
Ra-225				9.1E-1
Ra-226				4.6E-1
Ac-225				9.1E-1
Th-229				9.1E-1
Th-230				4.6E-1
Th-234				3.2E-1
Pa-233				8.5E-1
Pa-234				3.2E-1
U-233				9.1E-1
U-234				4.2E-1
U-236				3.9E-1
U-238				3.2E-1
Np-237				8.5E-1
Np-239			6.7E0	
Pu-238		1.1E3		
Pu-239			2.4E2	
Pu-240		5.3E2	1.8E2	
Pu-241		1.0E3		
Pu-242				2.9E-1
Am-241		3.8E3		
TOTALS	2.2E6	4.0E4	4.3E2	2.0E1

* Radionuclides constituting 1.0% or more of total radioactivity at indicated post-irradiation time.

FIGURE 1

RELATIVE HAZARD OF SPENT NUCLEAR FUEL (33,000 MWD BURNUP) vs. EQUIVALENT QUANTITY OF URANIUM ORE



NUCFUEL3P.A01/1-19-89

FIGURE 2

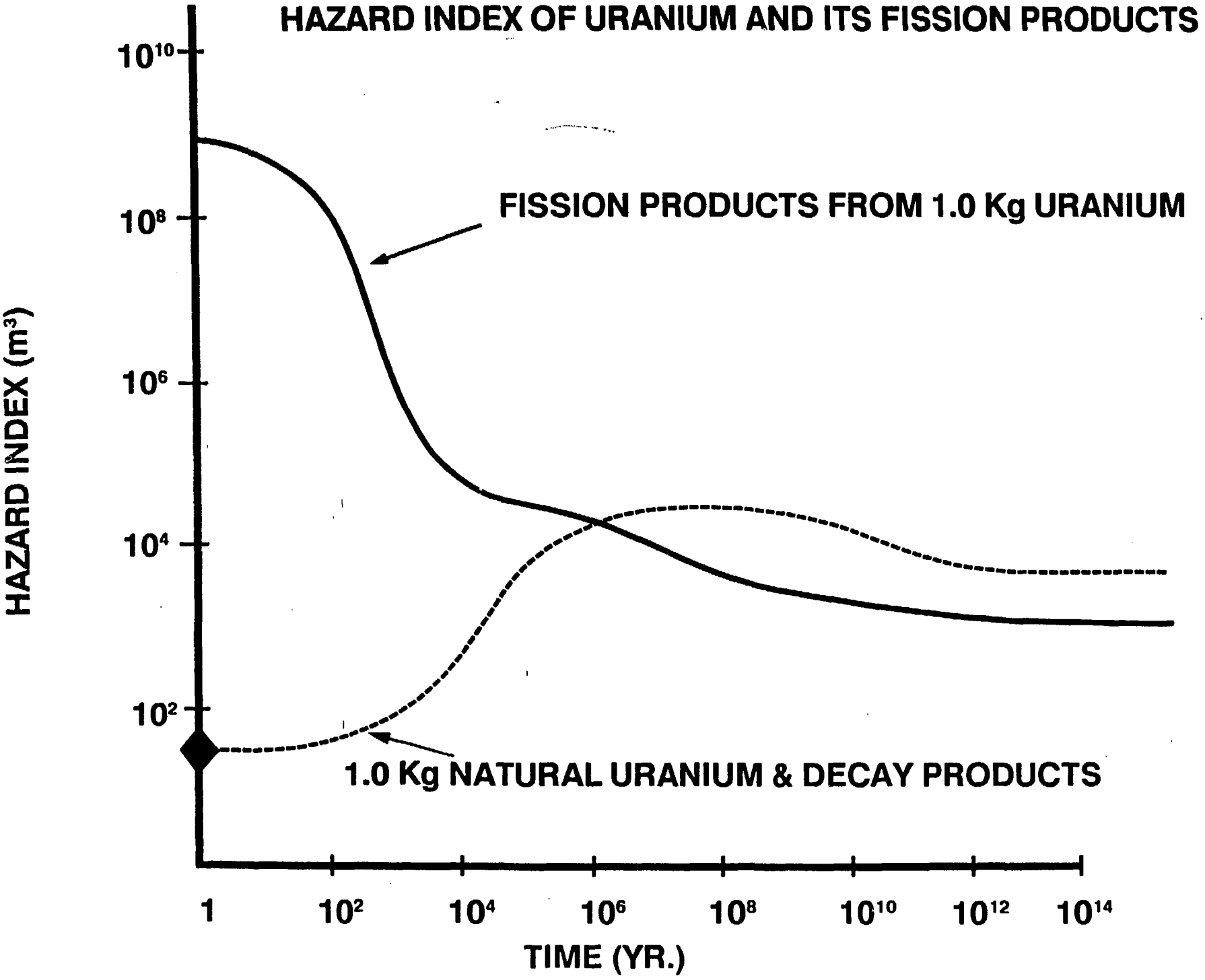

HAZARD INDEX OF URANIUM AND ITS FISSION PRODUCTS
FISSION PRODUCTS FROM 1.0 Kg URANIUM
1.0 Kg NATURAL URANIUM & DECAY PRODUCTS
HAZARD INDEX (m³)
10¹⁰
10⁸
10⁶
10⁴
10²
1
10²
10⁴
10⁶
10⁸
10¹⁰
10¹²
10¹⁴
TIME (YR.)

FIGURE 3

RELATIVE HAZARD OF SPENT NUCLEAR FUEL (33,000 MWD BURNUP) vs. EQUIVALENT QUANTITY OF URANIUM ORE (BASED ON ICRP-48)

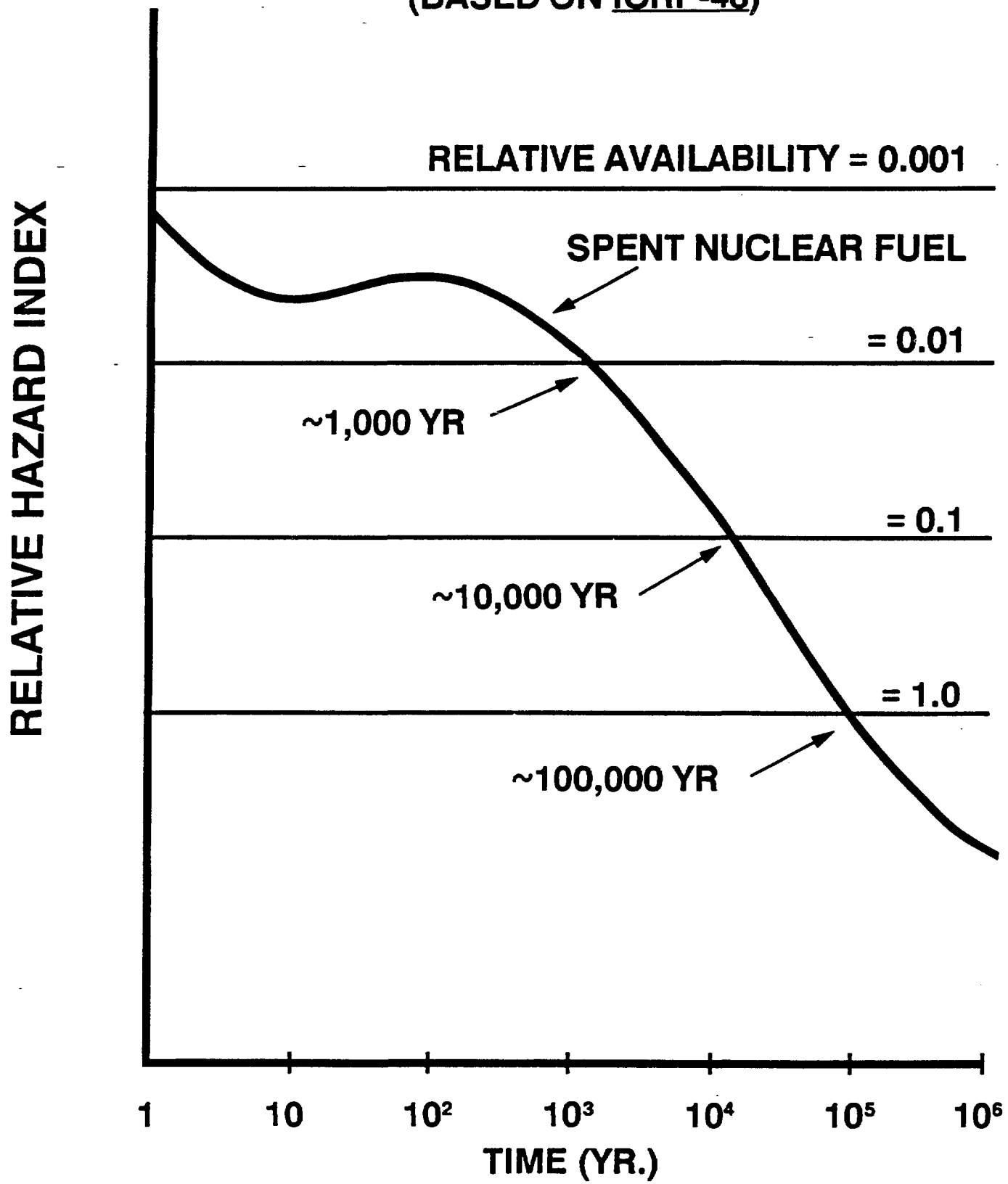


FIGURE 4

NATURAL AND ENGINEERED WASTE REPOSITORY BARRIERS

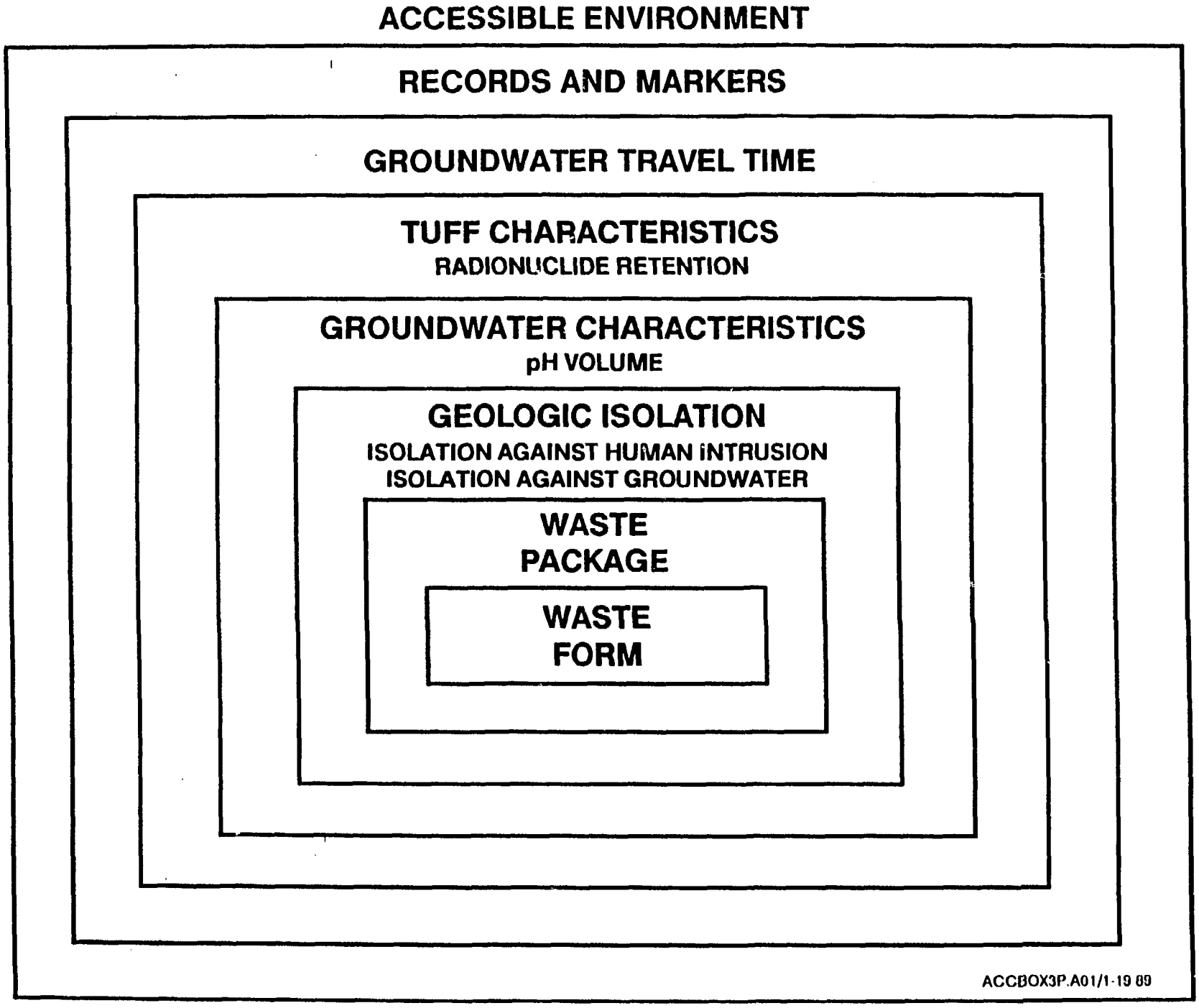
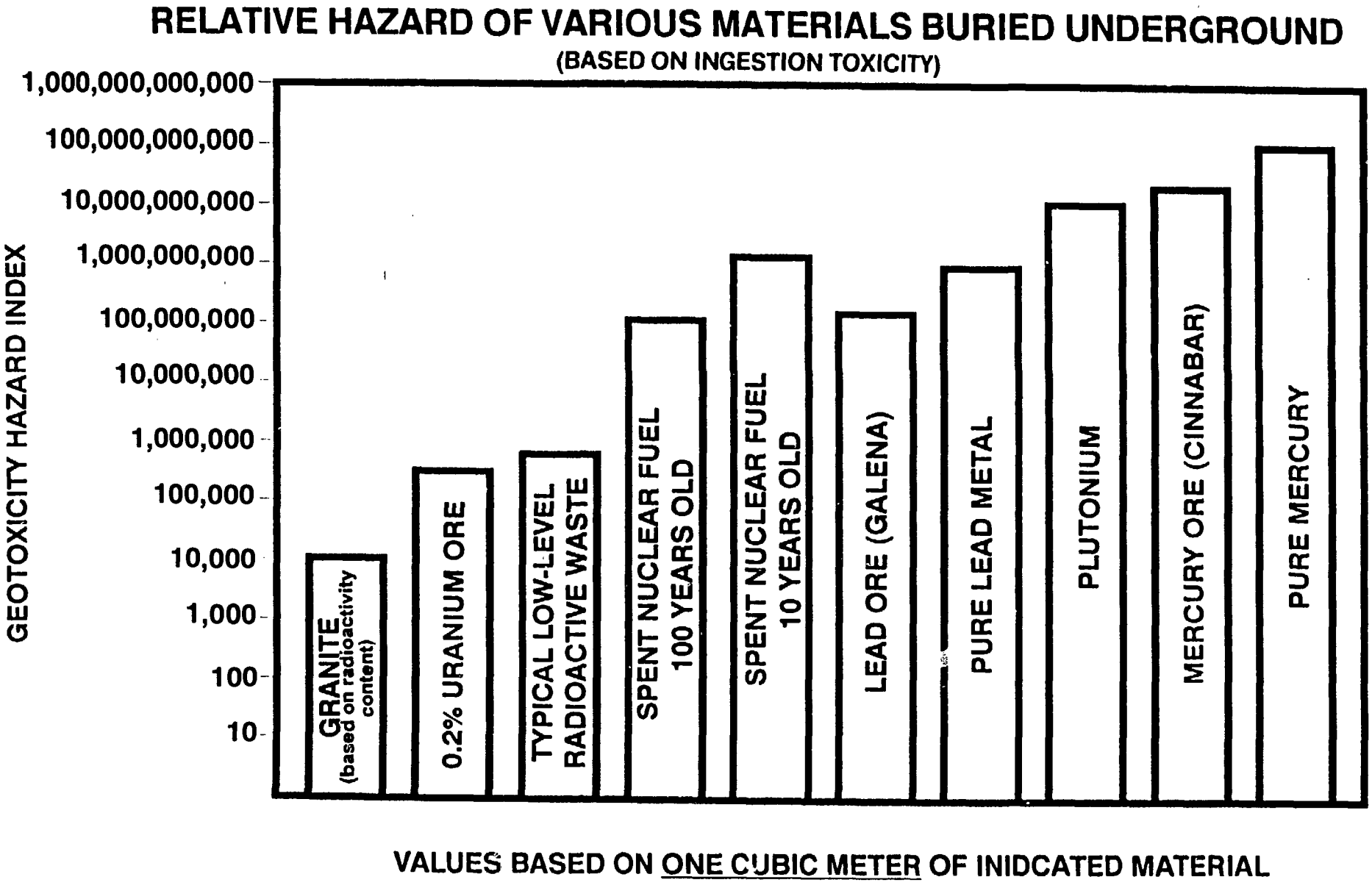


FIGURE 5


RELATIVE HAZARD OF VARIOUS MATERIALS BURIED UNDERGROUND
(BASED ON INGESTION TOXICITY)
GEOTOXICITY HAZARD INDEX
1,000,000,000,000
100,000,000,000
10,000,000,000
1,000,000,000
100,000,000
10,000,000
1,000,000
100,000
10,000
1,000
100
10
GRANITE (based on radioactivity content)
0.2% URANIUM ORE
TYPICAL LOW-LEVEL RADIOACTIVE WASTE
SPENT NUCLEAR FUEL 100 YEARS OLD
SPENT NUCLEAR FUEL 10 YEARS OLD
LEAD ORE (GALENA)
PURE LEAD METAL
PLUTONIUM
MERCURY ORE (CINNABAR)
PURE MERCURY
VALUES BASED ON ONE CUBIC METER OF INIDCATED MATERIAL