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**SURVEY OF NATURALLY OCCURRING HAZARDOUS
MATERIALS IN DEEP GEOLOGIC FORMATIONS:
A PERSPECTIVE ON THE RELATIVE HAZARD
OF DEEP BURIAL OF NUCLEAR WASTES**

MASTER

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Contents

Abstract.	1
Introduction	1
Analysis	2
Comparison of Hazard.	9
Conclusion	12
References	13

SURVEY OF NATURALLY OCCURRING HAZARDOUS MATERIALS IN DEEP GEOLOGIC FORMATIONS: A PERSPECTIVE ON THE RELATIVE HAZARD OF DEEP BURIAL OF NUCLEAR WASTES

Abstract

Hazards associated with deep burial of solidified nuclear waste are considered with reference to toxic elements in naturally occurring ore deposits. This problem is put into perspective by relating the hazard of a radioactive waste repository to that of naturally occurring geologic formations. The basis for comparison derives from a consideration of safe drinking water levels. Calculations for relative toxicity of fast breeder reactor (FBR) waste and light water reactor (LWR) waste in an underground repository are compared with the relative toxicity indices obtained for average concentration ore deposits. Results indicate that, over time, nuclear waste toxicity decreases to levels below those of naturally occurring hazardous materials.

Introduction

Current discussion concerning disposal of hazardous high-level nuclear wastes often centers on the possibility of deep burial of the solidified waste in geologic formations. The principal hazard from such a practice would be the consequence of escape to the biosphere of leached radiotoxic material into water supplies.

To place these hazards in perspective with other risks to society, it may be beneficial to consider the health effects from release of naturally occurring toxic materials in

geologic deposits. Concern has been expressed with regard to the extreme longevity of nuclear waste products, particularly the transuranic elements. Plutonium-239, for example, has a half-life of 24,000 years. Therefore, the toxicity and potential hazard from these wastes will persist for a long time. The hazard associated with certain toxic, stable elements must be considered in terms of eons, since the beginning of geologic time. The potential for contamination of surface water supplies by these hazardous minerals has always been

present, depending on leach rates in underground deposits, and more recently as a result of mining and refining activity. Indeed, many occurrences of heavy metal concentrations in surface waters have their origins in geologic formations within the watershed areas. This fact has been valuable in prospecting for ores.

A discussion of these ores will relate to certain toxic elements and radionuclides for which standards have been set to protect drinking

water supplies. These elements include: arsenic, barium, cadmium, chromium, mercury, lead, selenium, silver, thorium, and uranium. In the context of potential hazard resulting from toxic materials in geologic formations or from buried nuclear waste, the primary pathway for biological exposure is by ingestion. Therefore, the hazard resulting from the concentration of toxic materials in water supplies should provide the most rational basis for comparison.

Analysis

To evaluate the hazardous nature of these naturally occurring elements, two approaches were used:

- Determination of the crustal abundance of the mineral in the lithosphere and then calculation of the amount of water needed to dissolve the entire inventory of the mineral to bring its concentration in the resulting solution down to either safe drinking water standards or maximum permissible concentration levels as determined for radioactive substances in water (MPC_w) (see Table 1 and Fig. 1),
- Calculation of the concentration of these toxic elements in natural ore deposits and

further analysis of the amount of water necessary to dilute one cubic meter of ore down to the concentration specified in safe drinking water standards (Tables 2-4). Using low-grade uranium ore (0.2% ^{238}U) as a standard, the "toxicity indices" (T.I.) were calculated (in a way similar to that formulated by Haug⁸) for each of the toxic elements. This toxicity index allows not only for comparison of relative toxicities between naturally occurring elements, but also allows for comparison of naturally occurring hazardous minerals with nuclear waste material stored in an underground repository (Fig. 2).

Table 1. Toxicity index for certain naturally occurring toxic minerals in the Earth's lithosphere.

Element ^a	Crustal abundance, ^b ppm	Crustal mass, ^c g	DWC, ^d mg/liter	Water Dilution, ^e ml	T.I. ^f
As	5.0	1.0×10^{20}	0.05	2.0×10^{27}	10^5
Ba	430.0	8.6×10^{21}	1.0	8.6×10^{27}	4.3×10^5
Cd	0.18	3.6×10^{18}	0.01	3.6×10^{26}	4.3×10^4
Cr	200.0	4.0×10^{21}	0.05	8.0×10^{28}	4.0×10^6
Pb	16.0	3.2×10^{20}	0.05	6.4×10^{27}	3.2×10^5
Hg	0.5	1.0×10^{19}	0.002	5.0×10^{27}	2.4×10^5
Se	0.09	1.8×10^{18}	0.01	1.8×10^{26}	9.0×10^3
Ag	0.02	4.0×10^{17}	0.05	8.0×10^{24}	4.0×10^2
Radio-nuclide ^g			MPC ^d $\mu\text{Ci}/\text{ml}$		
^{235}U	0.014	2.8×10^{17}	3.0×10^{-5}	2.0×10^{22}	1.0
^{238}U	2.0	4.0×10^{19}	4.0×10^{-5}	3.35×10^{23}	1.7×10^1
^{232}Th	7.0	1.4×10^{20}	1.0×10^{-6}	1.54×10^{25}	7.7×10^2

^aInorganic elements for which EPA primary interim drinking water standards exist.

^bPercentage of the element found in the lithosphere (defined as being 5-36 km thick with a density of 2.6-2.9 g/cm³).^{2,3}

^cGrams of the element found in the lithosphere based on a conversion factor of 20×10^{24} g total for the entire lithosphere.²

^dMaximum permissible concentrations of the element or the radionuclide in drinking water, the former being in terms of mg/liter (drinking water concentration/DWC) and the latter in $\mu\text{Ci}/\text{ml}$ (maximum permissible concentration/MPC).^{1,4}

^eCalculated volume of water needed to dilute the total concentration of the given element in the lithosphere to safe drinking water standards.

^fRelative toxicity index ($^{235}\text{U} = 1.0$).

^gNaturally occurring radionuclides having the following activity levels: $^{235}\text{U} = 2.1 \mu\text{Ci}/\text{g}$; $^{238}\text{U} = 0.335 \mu\text{Ci}/\text{g}$; $^{232}\text{Th} = 0.11 \mu\text{Ci}/\text{g}$.⁵

The standards set by the Environmental Protection Agency (EPA)¹ and the National Council on Radiation Protection (NCRP)⁴ form the basis

for an analysis of the comparative toxicity of these elements in terms of crustal abundance, dilution ratios, elemental composition of *in situ*

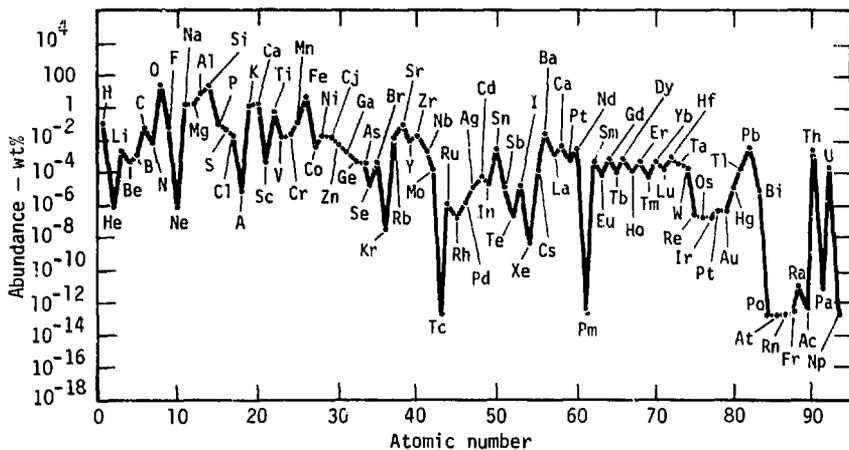


Fig. 1. Crustal abundance of the chemical elements.⁶

Table 2. Naturally occurring elements and their percentage in ores.⁷

Element	Ore	Maximum %, compound	Maximum %, element	Average %, compound	Average %, element	Chemical form
As	Sulphur-arsenides	1.9%	1.4%	0.2%	0.15%	As ₂ O ₃
Ba	Barite; Witherite	98.0%	57.0%	25.0%	14.7%	BaSO ₄
Cd	Associated with ZnS in sphalerite	4.43%	3.44%	1.0%	0.8%	CdS
Cr	Chromite	48.0%	32.8%	35.0%	24.0%	Cr ₂ O ₃
Pb	Pb-Zn ore/ galena	16.8%	14.5%	6.7%	5.8%	PbS
Hg	Cinnabar	25.0%	21.6%	3.0%	2.6%	HgS
Ag	Au-Ag ore	—	1.0%	—	0.1%	—
Se	Sulphides	—	6.3%	—	2.0%	—
²³⁸ U	Pitchblende	85.0%	72.0%	—	0.2%	U ₃ O ₈

Table 3. Dilution ratios calculated for maximum concentration of the element in its mineral ore with reference to safe drinking water standards (DWC).

Element	Maximum % of element in ore	Concentration of element, ore, g/m ³	DWC, mg/liter	Dilution ratio	T.I. ^a
As	1.4%	3.5×10^4	0.05	7.0×10^5	9.9
Ba	57.0%	1.43×10^6	1.0	1.43×10^6	2.0×10^1
Cd	3.44%	8.59×10^4	0.01	8.59×10^6	1.21×10^2
Cr	32.8%	8.2×10^5	0.05	1.64×10^7	2.31×10^2
Pb	14.5%	3.63×10^5	0.05	7.3×10^6	1.0×10^2
Hg	21.6%	5.4×10^5	0.002	2.7×10^8	3.8×10^4
Ag	1.0%	2.5×10^4	0.05	5.0×10^5	7.0
Se	6.3%	1.6×10^5	0.01	1.6×10^7	2.25×10^2
²³⁸ U	72.0%	1.8×10^6	0.07	2.6×10^7	3.66×10^2
²³⁸ U ^b	0.2%	5.0×10^3	0.07	7.1×10^4	1

^aToxicity index.

^bBase level, low grade uranium ore.

Table 4. Dilution ratios calculated for average concentration of the element in its mineral ore with reference to safe drinking water standards (DWC).

Element	Average % of element in ore	Concentration of element in ore, g/m ³	DWC, mg/liter	Dilution ratio	T.I. ^a
As	0.15%	3.75×10^3	0.05	7.5×10^4	1.056
Ba	14.7%	3.68×10^5	1.0	3.68×10^5	5.18
Cd	0.8%	2.0×10^4	0.01	2.0×10^6	2.8×10^1
Cr	24.0%	6.0×10^5	0.05	1.2×10^7	1.7×10^2
Pb	5.8%	1.45×10^5	0.05	2.9×10^6	4.0×10^1
Hg	2.6%	6.5×10^4	0.002	3.25×10^7	4.6×10^2
Ag	0.1%	2.5×10^4	0.05	5.0×10^5	7.04
Se	2.0%	5.0×10^4	0.01	5.0×10^6	7.0×10^1
²³⁸ U ^b	0.2%	5.0×10^3	0.07	7.1×10^4	1

^aToxicity index.

^bBase level, low grade uranium ore.

ores, and toxicity indices. The rationale for such comparison of radiologic and heavy metal hazards requires further elaboration.⁹

Toxicity is usually expressed in terms of minimum lethal dose (MLD) or LD₅₀ (amount of the substance which will kill 50% of the exposed

subjects in a specified amount of time). However, these measures do not take into account the effect on the organism of chronic low-level exposure to a certain chemical agent. To protect the general public from potentially harmful doses of deleterious elements or radiation, the EPA

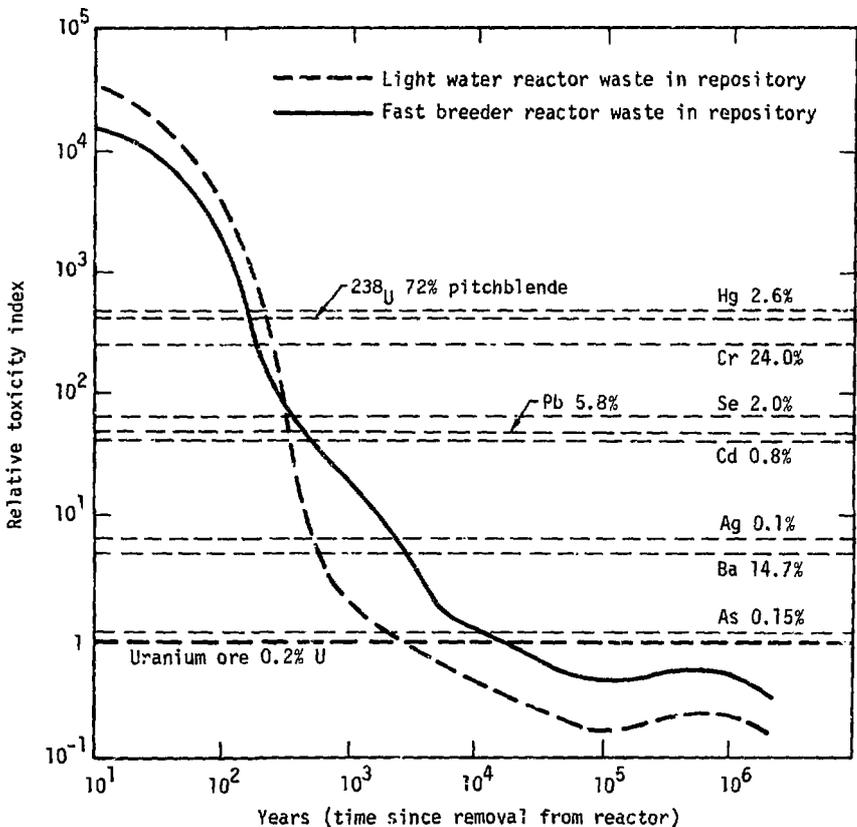


Fig. 2. Relative toxicity of nuclear waste over time, compared with that of average mineral ores of toxic elements.

and NCRP have devised standards for permissible concentrations of certain elements and radionuclides in drinking water. The premise for these standards is that continual exposure of the general public to these low levels of contamination of drinking water will not entail undue risk to human health. These standards have been formulated by scientific judgment based on a pooling of data on the toxic properties of the elements.

The setting of the EPA standards for drinking water has proceeded so slowly that only nine elements have been listed thus far.¹ However, even with definitive standards, the question of the hazard of exposure to low levels of certain elements is still open to question. The standards devised by the NCRP for maximum permissible levels of radioactivity in drinking water were formulated before the EPA heavy metal standards. Pursuant to the Atomic Energy Act of 1954, standards were set for levels of radioactivity in terms of Ci/ml of air and water⁴. These levels were calculated to account for the effects of such radiation levels on both genetic and somatic aspects of human health.

Hazardous substances in the earth's crust predate man, but he has evolved in their presence. Earlier generations of man have been exposed to even higher geologic amounts of

such radioactive species as uranium, thorium, and radium. Crustal abundances of specific hazardous elements have been expressed in terms of ppm (parts per million) and converted to grams:

$$\text{ppm} = \frac{\text{mass of the toxic element}}{\text{crustal mass}} \times 10^6$$

The dilution volumes obtained in the course of this analysis represent the amount of water that would be needed to dilute the total crustal mass of a specific element down to levels considered acceptable by EPA and NCRP. As an absolute number, this dilution factor has no meaning. Using that nuclide requiring the least amount of water for dilution to safe levels as a standard, ²³⁵U, a dilution ratio or toxicity index (T.I.) was calculated for each of the 11 elements and radionuclides in Table 1. This comparative measure indicates levels of hazard with reference to other naturally occurring substances. Both relative toxicity and abundance are incorporated into this index. Therefore, it is not surprising that the naturally occurring radionuclides have the smaller relative toxicity indices. This exercise is therefore useful in demonstrating the potential hazards of the natural environment -- hazards which the public routinely accepts, or at least tolerates, even

though they are several orders of magnitude greater than hazards posed by *in situ* radioactive ores. This comparison is particularly apt when we consider naturally occurring ore deposits of heavy metals in relation to proposed nuclear waste repositories. Ore formations are actively sought and penetrated for exploitation, whereas waste repositories would be generally avoided by man, thus resulting in a lesser chance of being breached to release toxic material to the environment.

A method of assessing relative geologic hazard without having to consider absolute masses of the elements in the lithosphere was arrived at through the calculation of dilution ratios (D.R.) for naturally occurring ores. For this analysis the *in situ* ores are assumed to be soluble in water.

The original assumption incorporated in the calculation of the toxicity index for the elements and nuclides included in this analysis concerns average ore density, which is considered to be equivalent to the average crustal density, or 2.5 g/cm^3 . In the calculations, the mass of the element under consideration per cubic meter of ore has been determined based on both the maximum percent and the average percentage of the element in its characteristic ore (see Tables 2-4, (i.e., ^{238}U in

pitchblende or Hg in cinnabar). The dilution ratio was arrived at by taking into account this calculated concentration in g/m^3 , and the drinking water standards set by the EPA in mg/liter. The drinking water standard for naturally occurring uranium was calculated to be 0.07 mg/liter based on Claiborne's¹⁰ hazard index for this toxic material. This standard takes into account the radiotoxicity of ^{238}U and its daughter products. The dilution ratio represents the relative amount of water required to dilute a cubic meter of ore to levels that will meet regulations to protect the public from undue radiation exposure or excessive ingestion of toxic, heavy metals. Finally, the toxicity index for each element was determined by using 0.2% uranium ore as a standard.

Once this toxicity index had been determined, it was compared with the toxicity indices devised by Haug for solidified reactor waste placed in an underground repository.⁸ A comparison of relative hazard is shown (Fig. 2) for average percentage of the element in its ore. Previous comparisons had been made between nuclear reactor wastes and 0.2% ^{238}U in low-grade pitchblende. This graphical analysis is an extension of Haug's comparison.

In his analysis Haug determined the radiotoxicity of a nuclide mixture

from a light water reactor (LWR) and a fast breeder reactor (FBR) and plotted these toxicity indices over time (as the waste decayed). Inherent in his analysis are the following assumptions:

- MPC_w's for the transuranic elements have been determined by comparison with NCRP standards for ²²⁶Ra.
- The radiotoxic waste products are considered to be present in a solidified matrix of low solubility.
- These radioactive waste products are to be located in single bore holes in deep geologic formations (cylinders 20 cm diam located in bore holes 10 m apart).

- The graphical comparison is made between the radiotoxicity of the waste cylinders along with the surrounding rock, and the radiotoxicity of the same volume of low-grade ²³⁸U ore (0.2%).

Haug's Relative Toxicity Index (RTI) is a dimensionless measure of hazard based on the radioactivity of 0.2% uranium ore diluted to safe drinking water standards. It was calculated in a manner similar to the toxicity index (T.I.) in this consideration of naturally occurring hazardous materials. Therefore, it was considered valid to superimpose the heavy metal element toxicity indices on Haug's original graph showing the toxicity of LWR and FBR wastes over time.

Comparison of Hazard

These calculated dilution ratio-toxicity indices serve to relate actual ores in geologic formations and their relative hazard should they be released. The dilution ratio-toxicity indices for the richest ore of a given element (Table 3) indicate the potential hazard to the public in the event of complete dissolving of the ore in drinking water. The predominant fear concerning buried nuclear waste is the potential for its dilution in ground water supplies, which might lead to ingestion by

humans. The ratios calculated for average ore concentrations (Table 4) appear to represent a more reasonable figure for potential risk. However, consideration of these two ratios indicates no more than one order of magnitude difference in dilution ratios. Therefore, use of either average or rich ore in this analysis appears to be appropriate. A simple comparison of the calculated ratios may be made to assess the relative hazard to human health from these naturally occurring toxic ores.

Radioactive ore considered in the analysis (0.2% U) requires considerably less dilution to reach safe levels for ingestion compared with certain heavy metal ores.

These dilution ratios may then be considered with reference to those volumes of water needed to dilute solidified nuclear waste in underground repositories to "permissible" levels. This information is provided by Haug in his graphical presentation of calculated toxicity indices over time for low-grade uranium ore and nuclear reactor wastes (Fig. 2).

Using his original graph, with 0.2% uranium ore as the baseline, Haug concludes that after 1000 years of decay, the actinides in the waste will have a radiotoxicity approximately equal to that of low-grade uranium ore.⁸ It has further been argued, notably by B. L. Cohen,¹¹ that mining uranium and then replacing it with high-level wastes might actually decrease the net hazard from radioactive material in the geosphere, when viewed over several centuries.

Adding to Haug's original graph the calculated toxicity indices for some of the toxic ores points up the relative hazard from material regularly brought to the surface for commercial use.

When calculated toxicity indices for the elements cited are compared with similar indices calculated for

nuclear waste, some interesting results follow. Deep geologic deposits of some of the most important industrial ores (lead, mercury) may be more potentially hazardous to human health than the proposed "deposits" of deeply buried solidified nuclear waste.

A further dimension may be added to this comparison when we consider that nuclear waste emplaced in stable geologic formations (such as granite or salt domes) would decay to a non-radioactive form over time, while the toxic metals maintain their specified level of toxicity infinitely. For example, several lead isotopes have no measurable half life.

These naturally occurring hazardous materials do not necessarily exist in stable deposits with limited access to the surface. Surface monazite sands characteristic of Brazil and Sri Lanka are known to contain high percentages of radioactive thorium.⁷ A recent instance of natural surface water contamination by radioactive material was discovered at Walker Lake, western Nevada.¹² Because of proximity to uranium deposits, the lake was found to contain approximately 130 parts per billion (ppb) uranium (600 short tons) which is 30 times higher than uranium concentration in sea water and about 130 times greater than that of fresh surface waters.

Contamination of drinking water in areas of toxic ore concentrations, such as the Almaden cinnabar mines of Spain, and the high-grade uranium mines of New Mexico and Utah, is but one of the surface manifestations of the man-mediated release of massive amounts of these toxic materials.¹³ Rather than exposing more of the deep deposits of these toxic metals through mining activities, the nuclear wastes emplaced in stable geologic deposits would likely be protected and left undisturbed by man's activity, since they would probably have little or no economic value.

In terms of this analysis, several assumptions need to be discussed. The numerical calculations are premised on the following:

- Crustal abundance figures are reasonable estimates of actual elemental composition of the lithosphere.
- The assumption of solubility of radionuclides and heavy metals *in situ* is similar to the solubility of the solidified nuclear waste matrix.
- The NCRP radioactivity standards (MPC_w) can be compared with EPA safe drinking water standards.

Composition of the constituent ores was obtained from descriptions of known commercial deposits of the metals being considered.^{7,14} Vein

deposits of some minerals may seem to contain anomalously high concentrations of these elements, but their existence has been documented. The "average" ore figures most definitely are subjective measures, but are the best that could be arrived at given commercial estimates of ore grades.

While this comparison between nuclear wastes deposited in deep geologic formations, and concentrations of potentially toxic minerals buried naturally in deep formations, might not be considered as apt by some, the location and accessibility of the minerals appear similar to possible sites for buried radioactive wastes. Thus, this analytical approach is justified.

For consistency, it is assumed that all of the elements or compounds in the ore in question are soluble in water. This assumption is acceptable when we consider the relative insolubility of most proposed solidified waste forms, those which have been recommended for emplacement in deep geologic formations. Indeed, great effort and expense will be invested in attempting to minimize the solubility of these waste forms (i.e., borosilicate glass, calcine, metal matrix).

For the crustal analysis (Table 1), it is necessary to consider drinking water standards promulgated by EPA as equivalent to NCRP standards for

maximum permissible concentrations of radioactive isotopes in water (DWC and MPC_w). This assumption might appear incongruous because of the comparison that is made between radioactivity levels ($\mu\text{Ci/liter}$) and elemental concentration (mg/liter). However, both standards may be considered minimum

levels of what constitutes a hazard to man. The formulation of these standards indicates that they were based on relatively the same criteria. Although the two sets of standards are not rigorous, they can be compared because they were formulated from the same premises.

Conclusion

This survey of naturally occurring hazardous substances in geologic formations is not an attempt to demonstrate the validity or acceptability of any type of waste disposal. Rather, it merely puts into perspective the hazards from geologic sources. The issue of disposal of high-level nuclear wastes will be resolved through both technical and sociological input. Individual attitudes may affect the final disposal plan. This

discussion of naturally occurring hazards already in geologic formations may increase awareness of the hazardous elements which the public routinely accepts because of their familiarity, ubiquitous nature, and commercial value. Assuming harm or hazard to man to be the overriding consideration in the establishment of a waste disposal methodology, then the comparisons made here could prove useful.

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