CALCULATING THE CHEMICAL HAZARD OF RADIOACTIVE WASTE

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ABSTRACT

Low-level radioactive wastes contain a wide variety of substances that are both radioactive and chemically hazardous. While the radioactive characteristics of these wastes have been defined, chemical components have been ignored from a management and regulatory point of view. In this paper, a methodology for comparing the hazards of various chemicals quantitatively with those of radioactive materials is presented in the form of a Waste Classification System. The system considers the hazards of materials based on intrinsic toxicity, persistence through time, availability to a human receptor, and buildup of decay or degradation products. The system has been applied to a number of materials to show its versatility. Examples are provided.

INTRODUCTION

Low-level radioactive waste contains a wide variety of substances that are both radioactive and chemically hazardous. While the radioactive characteristics of these wastes have been defined, chemical components have been ignored from a management and regulatory point of view. In this paper, a methodology for comparing the hazards of various chemicals quantitatively with those of radioactive materials is presented and examples of its versatility and use are provided.

Although chemical and radiological materials represent different kinds of hazards to a human receptor, there are similarities that can be used to compare these materials. For example, the longevity or persistence of a material, whether it has a half-life of a few seconds or will retain its toxicity indefinitely, can be quantified for either type of substance. Wastes, whether chemically hazardous or radioactive, can be physically stabilized in specific waste forms and then containers. All materials will eventually be transported through one or more pathways to be inhaled, ingested, or present an external hazard to a human receptor.

METHODOLOGY

These principles are shown in Equations 1 and 2:

$$\text{Hazard Index} = \text{Toxicity} \times \text{Persistence} \times \text{Available Hazard} \times \text{Progeny, or} \qquad (1)$$

$$HI = \frac{Q \times DCF}{DC} \times (P) \times (A \times WC \times WF) \times (PR) \qquad (2)$$

where

Q = the initial concentration (Ci/m^3 or mg/m^3)
DCF = the dose conversion factor (dose/unit intake)
DC = selected regulatory dose criteria (dose/yr)
P = persistence (unitless)
A = availability (m^3/yr)
WC = waste container factor (unitless)
WF = waste form factor (unitless)
PR = progeny factor (unitless).

A hazard index is calculated for each radionuclide and chemical in the waste, for each exposure pathway (ingestion, inhalation, and external exposure). The hazard indexes are summed for a total hazard, and a Waste Classification Index[1] is calculated:

$$WCI = \log_{10} \sum_{n=1}^{k} HI_n \qquad (3)$$

where

WCI = Waste Classification Index (unitless)
k = the total number of toxic materials in the waste
HI = hazard index.

Initial Concentration

The initial concentration is the estimated concentration of a radionuclide or chemical in the waste. In general, the concentration would be estimated in individual waste containers, although application of the formula to entire waste shipments or disposal areas is also possible.

Dose Conversion Factor[2]

The dose conversion factor (DCF) for ingestion and inhalation pathways is included in the equation to convert intake to dose. For nonradioactive substances, DCF is equal to 1, since the dose criteria (below) are in mass units. The dose conversion factor gives the 50-year dose commitment that results per unit of radionuclide ingested or inhaled.[3] Organ doses are weighted according to organ radiosensitivity, and then summed to give an effective whole-body dose. The effective whole-body dose is intended to represent the same risk as if the entire body were irradiated uniformly. For external exposure, the DCF is combined with the availability term and is applicable to radiological substances only. Exposure rates for gamma-emitting radionuclides have been measured 1 m above soil containing homogeneously mixed, known quantities of radionuclides.[4] Exposure rates from other radionuclides were extrapolated, based on photon energies and branching ratios.

Dose Criteria[2]

Dose criteria are used to equate the effects of radiological and chemical waste substances on a similar scale. Summation of radiological and chemical indexes is valid only if the DC values are comparable.

Multimedia Environmental Goals (MEGs)[5] have been developed for the U.S. Environmental Protection Agency in an attempt to define levels of contaminants in effluents, emissions, and in the ambient environment (air, water, and land) appropriate to protect human populations and ecosystems. The subcategory of MEGs used in this analysis is a health-based system called Estimated Permissible Concentration (EPC).

The MEG methodology is, in theory, well suited for use as a dose criterion. Values are reported for a large number of chemicals. The health-based EPC values have been developed from a variety of sources including federal standards and criteria, threshold limit values (TLVs) and NIOSH recommendations, LD_{50} data, and indicators of genotoxicity or carcinogenic potential. The methodology uses simple, conservative models to derive values for various exposure routes (air, water, and land). Two major disadvantages are evident. First, a variety of models are used in the system, many of which are extremely simplistic and may result in EPC values varying by several orders of magnitude for a given substance. Second, little attempt is made to normalize values calculated through the various models. That is, the MEG methodology places heavy reliance on the use of TLVs to calculate EPC values not only for air, but for water and soil also.

In order to develop a unified index to evaluate both radioactive and chemical materials, both types of materials must have dose criteria that are developed from a similar methodology. The radiological dose criterion selected for use is 50 mrem/yr, derived from the occupational exposure limit of 5 rem/yr and a safety factor of 100. This is the same safety factor used in development of EPC values from TLVs.

Normalization of dose criteria is very difficult because of the different dose-response curves of many chemicals. The dose criteria selected and briefly described here are examples of how normalization may be attempted; at the present time, this appears to be the most workable system. Work is continuing to refine this particular parameter of the Waste Classification System.

Persistence[2]

The term persistence describes the longevity of a material over a specified period of time. In this study, the calculational period used is 500 years, the longest time period identified by the U.S. Nuclear Regulatory Commission that a low-level waste disposal facility must perform. One-hundred years may be a more appropriate time period.

For radioactive substances, longevity is a function of each nuclide's decay rate. Relatively short-lived nuclides such as Co-60 or H-3 do not persist over the 500 years and therefore have small persistence values. The longer-lived Pu-239 and U-238 have values of 1, indicating that essentially all the material will be available for 500 years and beyond.

Several methods for describing persistence have been investigated. For the example in this paper, P_n is a time-averaged fraction of the initial radionuclide concentration, or

$$P = \frac{1}{t}\int_0^t e^{-\lambda_n t}dt \qquad (4)$$

where

t = 500 years
λ_n = the radioactive decay constant in $year^{-1}$.

Describing the persistence of nonradiological materials presents different problems. Little decomposition will occur while the chemical waste materials are in containers protected from the environment. Hence, two half-life or decay constants, based on studies of reaction rates of chemical processes, are required to evaluate the persistence of chemicals.[6] The first decay constant evaluates the persistence inside the container (from time zero to container breach); the other evaluates persistence outside the container (from container breach to the end of the evaluation period, about 500 years).

A persistence factor of 1 is the reliably conservative option when data for decay constants are not available.

Availability

The availability factor describes the rate at which a substance can leave the soil and enter a transport pathway to man. Exposure modes considered in the system are ingestion, inhalation, and external exposure.

The specific approach used was an analog approach.[7,8] Analogs are naturally occurring materials that can be used to establish a reference availability for buried materials. Analog data are based on empirical observations of the behavior of natural materials, and circumvent the need for detailed environmental pathway modeling. For materials with natural analogs, direct data related to abundance and human intake rates were applied. For materials such as organics, surrogate analogs were developed using a benchmarking technique. Benchmarking is a systematic comparison of a series of fundamental properties of chemicals whose environmental behavior is unknown with a chemical standard whose environmental behavior is known.[9] For example, mercury is used as a surrogate for the inhalation of volatile organic and inorganic substances. Likewise, DDT is used as a surrogate for the ingestion of nonvolatile organic and inorganic substances. Both substances were selected because they are ubiquitous and occur in relatively stable concentrations in the environment. Table I shows the major exposure pathways and the type of analog used for various types of substances.

Two assumptions were used in the development of the availability factor.[8] First, general populations are the receptors of primary concern. Accordingly, availability to intruders or other definable maximum individuals is not included. Second, the availability factor is defined in terms of the fractional uptake for the various exposure modes (ingestion, inhalation) and/or the consequence of that exposure per unit quantity of buried material (i.e., the effects of external radiation exposure).

Waste Container Factor

The waste container factor is the first barrier to waste migration. During the time the container is intact, there is no release of waste and the waste safely decays or degrades in place. As soon as the container is breached or opened tc the surrounding soil, the waste may migrate to man.

The waste container (WC) factor is defined as the fraction of original waste material remaining when a container is breached; hence it accounts for the effect of the lifetime of the container on waste material concentration. The values of WC will vary between 0 and 1. When the container life is long and the material life short, the majority of the material will decay or degrade in the container without being exposed to the soil, and the WC value approaches 0. Likewise, when the container life is short and the material life is very long, then the value of WC approaches 1. For the example in this paper, the container life of a 55-gal steel drum is assumed to be six years[10] and $WC_n = e^{-\lambda} n\ (6\ yr)$. (5)

Waste Form Factor

The waste form (WF) is another barrier to potential waste migration. When a material is mixed with a fixation agent like concrete, the rate at which the material is released to the surrounding soils is greatly reduced. Waste forms such as loosely packaged paper trash do not inhibit release of waste materials; hence, contaminants can readily enter the soil once the waste container is breached. The waste form factor is defined as the maximum fraction of material that could be released from a container in the first year after the container is breached. WF estimates are based upon laboratory tests and may not reflect field conditions for solidified waste in soil. Also, since WF values have been calculated for only a small number of materials, using a waste form factor of 1 is the reliably conservative option at this time.

The terms availability, waste container factor, and waste form factor are individual parameters of the available hazard term. Availability describes the natural unencumbered movement of materials from buried waste, while the waste container and form factors are common engineered barriers "modifying" the availability factor. Other modification factors could be considered in site-specific analyses. Site factors such as geohydrology, burial depth, engineered factors (trench caps, etc.), and demography have the potential for greatly modifying availability. For this paper, however, a more generic analysis is called for, and site modification factors will not be examined.

Progeny

The term progeny is a calculated, unitless value designed to correct for the buildup of decay products. It involves modification of the availability and dose conversion factor and is specific for the radionuclides involved. A better discussion of this parameter can be found in Reference 11.

TABLE I
ANALOG APPROACHES USED

Class of Toxic Substances	Exposure Modes		
	Ingestion	Inhalation	Exposure
Radioactive			
Inorganic			
Volatile	N/A	A	A
Nonvolatile	A	A	A
Organic			
Volatile	N/A	S-Hg	A
Nonvolatile	S-DDT	A	A
Nonradioactive			
Inorganic			
Volatile	N/A	S-Hg	N/A
Nonvolatile	A	A	N/A
Organic			
Volatile	N/A	S-Hg	N/A
Nonvolatile	S-DDT	A	N/A

Abbreviations: N/A = not applicable; A = direct natural analog; S-Hg = surrogate analog using mercury; S-DDT = surrogate analog using DDT.

APPLICATION

Control rods from a pressurized water reactor, with a composition of 80% silver, 15% indium, and 5% cadium by weight, contain significant concentrations of silver and cadmium, both regulated by the EPA, such that the chemical hazard outweighs the radiation hazards. In addition, the stainless steel in the rod cladding and other stainless steel core components also presents a potential chemical hazard upon degradation. Table II contains hazard calculations for a hypothetical waste mixture containing, by volume, a 50% stainless steel--50% silver-indium-cadmium mixture, in a wooden box. The Waste Classification Index for this mixtures is

$$WCI = \log (5.1 \times 10^8) = 8.7. \quad (6)$$

This particular calculation takes no credit for either waste form or waste container; that is, both of these factors equal 1. A high-integrity container, or melting the metal components into a single ingot could greatly decrease the hazard of this package of waste.

Table III shows other hazard calculations on miscellaneous hazardous substances known to be contained in low-level waste.[12] These substances were selected to show the variety of chemicals that can be evaluated by the Waste Classification System. Amsco diluent, containing approximately 3% tributyl phosphate by volume, was assumed to fill 80% of a 55-gal drum. Benzene was assumed to fill 80% of a 5-gal drum. This 5-gal drum was probably overpacked into another container, but no credit for that was taken. For hafnium, it was assumed that 20 g were contained in a 55-gal drum. The Waste Classification Indexes of these materials ranged from 1.1 to 5.9. As a comparison, Ni-63, Sr-90, and Cs-137 had Waste Classification Indexes ranging from 1.1 to 1.6 for concentrations considered to be Class A waste according to the classification used by the NRC in 10 CFR 61. As with the chemical substances, no credit was taken for either a waste container or waste form.

TABLE II

HAZARD INDEX CALCULATIONS FOR CONTROL ROD MIXTURE[a,b]

		DCF_n		DC_n			A_n			
Substance	Q_n[c]	Ing[d]	Inh[d]	Ing[d]	Inh[d]	P_n	Ing[d]	Inh[d]	Ext[e]	HI[f] Total
C-14	2.6E-1[g]	2.1E+6	2.1E+6	50	50	9.7E-1	2.7	1.8E-7	0	2.9E4
Fe-55	2.2E+3	6.7E+6	2.6E+6	50	50	6.9E-3	6.5E-5	1.2E-7	3.5E-9	1.3E2
Ni-59	1.4	2.0E+5	1.3E+6	50	50	1.0	1.5E-3	2.2E-7	4.4E-4	8.4E0
Co-60	1.6E+3	2.6E+7	1.5E+8	50	50	1.5E-2	1.0E-2	2.4E-7	1.1E-3	1.2E5
Ni-63	2.1E+2	5.5E+5	3.1E+6	50	50	2.7E-1	1.5E-3	2.2E-7	0	9.4E2
Mn	8.0E+7	1.0	1.0	3.7E+1	1.0E+2	1.0	6.8E-4	9.9E-8	0	1.5E3
Fe	2.7E+9	1.0	1.0	2.6E+1	2.0E+1	1.0	6.5E-5	1.2E-7	0	6.8E3
Co	8.0E+6	1.0	1.0	1.1	1.0	1.0	1.0E-2	2.4E-7	0	1.9E0
Ni	3.6E+8	1.0	1.0	4.4E-1	3.4E-1	1.0	1.5E-3	2.2E-7	0	1.2E6
Cr	7.5E+8	1.0	1.0	2.2E-2	1.7E-2	1.0	3.9E-4	3.0E-7	0	1.3E7
Cd	2.5E+8	1.0	1.0	2.2E-1	1.7E-1	1.0	8.0E-2	4.8E-5	0	9.1E7
In	7.5E+8	1.0	1.0	2.6	2.0	1.0	2.7E-4	1.2E-5	0	8.3E4
Ag	4.0E+9	1.0	1.0	2.6	2.0	1.0	2.6E-1	3.5E-4	0	4.0E8
										5.1E8
Radiological units	Ci/m^3	mrem/Ci	mrem/Ci	mrem/yr	mrem/yr	--[h]	m^3/yr	m^3/yr	m^3/yr	
Chemical unit	mg/m^3	NA[i]	NA	mg/yr	mg/yr	--	m^3/yr	m^3/yr	NA[i]	--

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a. For calculational and comparison purposes, this waste mixture consists of 50% stainless steel and 50% elemental Ag-In-Cd mixture in a 1-m^3 volume wooden box. The hazard index of Nb-94 and C were negligible compared with other substances in the table and, hence, were omitted.

b. WF = 1 for all substances. This is a conservative assumption since the activation products in the stainless steel would not be as available for transport away from the waste as some other substances. Also because the waste is presumed to be packaged in a wooden box, WC = 1.

c. Concentrations of all radiological substances were gathered from Reference 3, Table 3.3 (L-FNRCOMP). The concentrations of the control rods were based on the rod elemental composition (80% Ag, 15% In, 5% Cd), density of the elements, and the amount of the mixture that would fit in half of a 1-m^3 box. The concentration of the stainless was based on element ratios and steel density from Reference 5, and the amount of the steel mixture that would fit in half of a 1-m^3 box.

d. Ing = Ingestion pathway, Inh = Inhalation pathway. The DCF for external exposure is combined with the A ext term; that is, A_n for external exposure = DCF x Ext.

e. Ext = external exposure pathway.

f. $HI_n = (Qn \times DCF_n/DC_n)\ (P_n)(A_n \times WC_n \times WF_n)(PR)$. HI_T is summed for all pathways; that is, $HI_T = HI_{ing} + HI_{inh} + HI_{ext}$.

g. 2.6E-1 = 2.6 x 10^{-1}.

h. Unitless value.

i. NA = not applicable.

TABLE III
HAZARD CALCULATIONS FOR MISCELLANEOUS HAZARDOUS SUBSTANCES

Substance	Concentration[a]	WCI[b]
Tributyl phosphate	1.3E9[c]	5.4
Benzene	3.5E9	5.9
Hafnium	9.6E4	1.1
Ni-63	3.5	1.2
Sr-90	0.04	1.1
Cs-137	1	1.6

a. Concentrations are in units of mg/m^3 for chemical substances and Ci/m^3 for radiological substances.

b. Waste container and waste form factors for all substances equal 1.

c. 1.3E9 = 1.3 x 10^9.

DISCUSSION

The waste classification system described in this paper is a tool to evaluate the relative hazards of chemical and radiological waste substances. For example, if a waste disposal site accepted waste with a classification of 5 or below, and a particular waste had a classification of 7, the index could be used to determine what parameters of the waste could be modified in order to decrease the hazard. The waste could be diluted (i.e., its concentration decreased by packaging in two containers); it could be treated to alter its waste form (incinerated, chemically processed, solidified into glass, cement, bitumen, etc.); it could be packaged into a better container; or perhaps site factors such as burial depth could be altered to decrease the hazard.

The chemical hazard of radioactive waste has been frequently neglected, although for some waste streams, the chemical hazard may be greater than the radiological hazard. Accurate characterization of chemical constituents, and classification of chemicals and radioactive materials by similar criteria are necessary to compare current regulations for different kinds of hazardous substances, as well as to develop new regulations pertaining to the management of toxic substances in radioactive waste. Such comparisons will highlight substances being over or underregulated and provide a basis for efficient utilization of our natural and economic resources.

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