Risk-Based Information to Support the Evaluation of Management Options for Cesium and Strontium Capsules at the Hanford Site - 8304

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ABSTRACT

Evaluations are under way to support U.S. Department of Energy decisions on how to manage cesium and strontium capsules currently in storage at the Hanford site. Health-based exposure limits for drinking water, oral toxicity data, and environmental fate information were combined in an initial evaluation to frame performance targets for managing chemicals and radionuclides that could leach from the capsules and migrate to groundwater over time. More than 50 relevant benchmarks were identified for 15 of the 17 contaminants in the study set. Of those multiple benchmarks, EPA limits for drinking water served as the main basis for the leachate performance targets. For the remaining two contaminants, stable cesium and zirconium, preliminary indicators were derived from a limited review of toxicity data. Thus, preliminary candidate concentrations were identified for the full study set to support the ongoing evaluation of capsule management options.

INTRODUCTION

The U.S. Department of Energy (DOE) is assessing long-term management options for nearly 2,000 radioactive capsules being stored at the Hanford site. Risk evaluations are under way to assess disposition alternatives for more than 1,300 cesium capsules and more than 600 strontium capsules in controlled storage. These assessments consider hypothetical scenarios that assume institutional controls are lost at some point in the future and people use underlying groundwater as a source of drinking water. Such assumptions help frame the evaluation of contaminants that could leach from the capsules over time. To support this assessment, health-based concentrations are needed to serve as performance targets for managing the capsules into the long term. These concentrations are to provide initial points of comparison for interpreting results of transport models being applied to assess capsule leachate under various conditions. An earlier scoping study of the projected leachate had identified eleven study contaminants: three radionuclides (Cs-135, Cs-137, and Sr-90) and eight chemicals, consisting of six more metals (barium, cadmium, stable cesium, chromium, lead, and silver) and two ions (chloride and fluoride).

APPROACH

The approach for developing health-based performance concentrations for leachate from the cesium and strontium capsules consisted of three main steps:

1. Determine whether relevant concentration or dose limits have been established for the study contaminants, assuming capsule leachate reaches groundwater and is used as drinking water over an extended future.

2. For relevant dose-based limits, derive target concentrations using standard exposure factors with the same assumptions regarding the future as above.

3. If neither concentration nor dose limits are found, review toxicity data and calculate a preliminary health-based indicator concentration.

Before implementing these steps, the study set was first checked to assess completeness by reviewing fate and toxicity characteristics.

For the radionuclides, this fate-toxicity evaluation involved first assessing whether any decay products should be added to the initial list that could contribute to radiological risk, to ensure that they were explicitly included in the radiological analyses. The relevant daughters are (1) Ba-137m, the short-lived decay product of Cs-137; and (2) Y-90, the short-lived decay product of Sr-90. Both were added to the study set.

Next, the three radionuclides were checked to determine whether chemical toxicity had been considered where appropriate. From this review, no additions were warranted for the two cesium isotopes because stable cesium was already included in the chemical list. In contrast, chemical toxicity is indicated for strontium, regardless of whether or not it is radioactive (e.g., Sr-90 or stable strontium). Because stable strontium was not on the initial study set, it was added to the list.

Third, the terminal decay products were checked to assess potential toxicity contributions, given the long time frame for capsule management. Cs-135 decays to stable Ba-135, Cs-137 to stable Ba-137, and Sr-90 to Zr-90. Stable barium is already included in the chemical list, so no addition was warranted for the two cesium isotopes. However, zirconium was not part of the original set so it was added to the chemical list.

The fourth check addressed the chemicals, with speciation being important for chromium because different benchmarks and toxicities have been identified for the hexavalent (oxidized) and trivalent (reduced) forms. Thus, in addition to addressing total chromium, it is useful to distinguish information for Cr VI and Cr III. Beyond this speciation consideration, the remaining chemicals on the preliminary list can combine with a number of others to form various compounds but do not themselves transform to other chemicals over time. Therefore, no other clarification or addition is warranted to assure coverage of fate products for the chemicals.

From these evaluations, the revised study set consists of five radionuclides and a dozen nonradioactive chemicals, with the six additions highlighted in italics in Table I.

Table I. Leachate Contaminants

Radionuclides		
1. Cs-135		
2. Cs-137	*3. Barium-137m*	
4. Sr-90	*5. Yttrium-90*	
Chemicals (Nonradioactive)		
1. Barium	5. Chromium, total	9. Lead
2. Cadmium	*6. Chromium III*	10. Silver
3. Cs-133	*7. Chromium VI*	*11. Strontium*
4. Chloride	8. Fluoride	*12. Zirconium-90*

In implementing the three main steps to identify target concentrations for the leachate, it is important to consider the nature of the toxicity information underlying existing benchmarks. Many standards and guidelines were established 10 to 20 years ago and our understanding of toxicity and biokinetics has improved considerably since then, as reflected in better data and models for estimating dose and risk. The emphasis is thus on values that have undergone rigorous technical review and reflect the most recent scientific knowledge.

For the chemicals in the study set, dose-based values were evaluated according to the hierarchy recommended by the U.S. Environmental Protection Agency [1], which prioritizes values from the EPA Integrated Risk Information System (IRIS) [2]. The second source is EPA provisional peer-reviewed toxicity values (PPRTVs). If those are unavailable, then the third source includes all other values, such as screening values from EPA regions (e.g., Region 9 [3]) and values from other federal, state, or international sources. These sources include the Food and Drug Administration (FDA) and World Health Organization (WHO).

For carcinogens, the EPA standard toxicity values reflect the increased probability (above a background rate) that an individual will develop cancer over a lifetime as a result of chronic exposures. This is also referred to as excess lifetime risk and is based on population statistics. To assess the noncarcinogenic toxicity of chemicals, the toxicity values in IRIS represents a daily "safe" level that is used to indicate whether a potential exists for an individual to incur an adverse effect from chronic daily exposures.

Calculation for Chemicals

Potential health effects from chemical exposures are estimated in two steps. First, an intake is calculated from the chemical concentration in the given medium (here, drinking water) combined with basic exposure assumptions (e.g., ingestion rate in liters/day [L/d] over a certain number of years [yr]). The intake is the amount taken in per unit body weight per unit time, generally expressed as milligrams per kilogram-d (mg/kg-d), and it is calculated using the following equation, with established default exposure values for a resident [4]:

$$I_i = \frac{C_i \times IR \times EF \times ED}{BW \times AT} \quad \text{(Eq.1)}$$

where:

I_i = intake of chemical *i* (mg/kg-d)
C_i = concentration of chemical *i* in water (mg/L)
IR = intake (ingestion) rate, assumed to be 2 L/d
EF = exposure frequency, assumed to be 365 d/yr
ED = exposure duration, assumed to be 30 yr
BW = body weight (kg), assumed 70 kg for adult
AT = averaging time (d): 10,950 d for noncancer effect; 25,550 d for cancer risk

A conversion factor (CF) is applied as warranted, for example to adjust mass units of micrograms (µg) to mg using the CF of 10^{-3} mg/µg. For drinking water, the daily exposure time (1 d) is included with the intake rate. For this study, the ED of 30 yr was used because it is the EPA default assumption for chronic residential exposures. While the EPA default value for EF under a residential scenario is 350 d/yr, to be further protective for this evaluation, the hypothetical individual is assumed to drink 2 L every day (365 d/yr) throughout the 30 yr. The calculated intake is then combined with the standard

toxicity value, which is both contaminant- and route-specific, to calculate the cancer risk or potential for noncancer effects.

Calculation for Radionuclides

The calculation for radionuclides follows the same basic concepts as for chemicals, with exposure expressed in terms of intake or dose in accordance with standard EPA guidance [4, 5, 6]. The intake can be represented by the amount of activity (picocurie [pCi]) taken into the body via ingestion, which is calculated using the following equation.

$$I_i = R_i \times IR \times EF \times ED \times CF \quad \text{(Eq. 2)}$$

where:

I_i = intake of radionuclide *i* (pCi)
R_i = concentration of radionuclide *i* in water (pCi/L)
CF = conversion factor (as indicated)

Similar to the approach for chemicals, the ingestion rate incorporates exposure time; IR, EF, and ED are as previously defined. The EPA has established standard coefficients to estimate cancer risk from ingestion of radionuclides, as given in Federal Guidance Report (FGR) 13 [5]. Cancer is generally considered the limiting effect for environmental radiological exposures [6]. (Note the chemical toxicity of selected radioisotopes is also addressed by considering the toxicity of the stable form, notably for strontium, following the approach indicated above.) The EPA radiological risk coefficients (RRCs) are expressed in units of risk per activity taken into the body and are age-and gender-averaged values. In FGR 13, the activity is given in the standard international unit of Becquerel (Bq). This is converted to pCi (which is commonly used in assessing risks at U.S. sites) by applying the factor of 1 Bq/27 pCi.

As described for the chemicals, the calculated intake is then combined with the radionuclide-specific risk coefficient for that route (ingestion) to estimate the cancer risk. The radiological risk coefficients are available to assess both cancer mortality and morbidity (incidence), both from tap water and overall dietary ingestion. For this project, the values for morbidity from ingesting tap water are used to frame the health-based concentrations identified for the capsule leachate.

RESULTS AND DISCUSSION

More than 70 information sources were reviewed to develop performance indicators for the 17 contaminants associated with leachate from the cesium and strontium capsules. These sources include drinking water standards from EPA and other organizations, as well as methodology updates and toxicity data from scientific reports and journals. From this review of existing concentration and dose limits alone, more than 50 concentrations were identified for the study chemicals.

Relevant benchmarks were found for all but two contaminants in the study set: stable cesium and zirconium. For example, seven values were identified for cadmium and eleven for fluoride, including several duplicates. This is not unexpected, given that when one organization establishes a value it is often adopted by others; alternately, a single study may be used by several organizations as the basis for deriving their benchmarks.

Selected results are highlighted in Table II. Calculated values are italicized. Limits identified by EPA for drinking water served as the main basis for the candidate concentrations for most of the contaminants.

Table II. Target Candidate Concentrations Identified for Capsule Leachate

Chemicals	*Concn (mg/L)*	*Basis and Context*
Barium	2	MCL (set in 1992); the goal (MCLG) is the same [7, 8].
	7	Calculated from RfD of 0.2 mg/kg-d for barium and compounds from a recent evaluation [2], using standard values of 70 kg, 2 L/d.
Cadmium	0.005	MCL (set in 1992); the MCLG is the same [7].
	0.018	For cadmium in water, calculated from RfD of 0.0005 mg/kg-d from 1994 [2] (a more recent evaluation than the MCL), assuming 70 kg and 2 L/d (rounded).
Cesium (stable)	800	No standard benchmark identified; this preliminary indicator was derived from limited toxicity data.
Chloride	250	Secondary MCL [7, 8], based on taste, not health effects.
	450	Calculated from the tolerable upper intake level of 3.6 g/d for adults (age 19 to 51+) [9]; assuming 25% from tap water, consistent with assumption for derived intervention levels [10], yields 450 mg/L.
Chromium, total	0.1	MCL (set in 1992); the MCLG is the same [7, 8].
Chromium VI	*0.11*	Calculated from RfD of 0.003 mg/kg-d [2] (from 1998, a more recent evaluation than the MCL), assuming 70 kg and 2 L/d (rounded).
Chromium III	*53*	Calculated from RfD of 1.5 mg/kg-d [2] (from 1998, a more recent evaluation than the MCL), assuming 70 kg and 2 L/d (rounded). Speciation data can be used to calculate the total based on toxicity; e.g., some data for Hanford areas with higher chromium VI suggest it may be 25-71% of total [11].
Fluoride	4	MCL (set >20 years ago, in 1986); the MCLG is the same, based on skeletal fluorosis [7, 8].
	2.1	Calculated from RfD of 0.06 mg/kg-d [2] (from 1989), derived from no-effect level (NOEL) for cosmetic effect (tooth discoloration) in children. Lowest observed adverse effect level (LOAEL) is 2 times higher, translating to 4 mg/L.
Lead	0.015	Treatment technique action level (if exceeded at the tap, a required process would be put in place, e.g., for corrosion control treatment) [8]. Toxicity-based blood level guidelines exist separately.
	0.01	Guideline value for drinking water [12].
Silver	0.2	Drinking water equivalent level (DWEL); also the 1-d and 10-d health advisory [8].
	0.18	Calculated from RfD of 0.005 mg/kg-d [2] based on a NOEL (from 1996), using standard default assumptions of 70 kg and 2 L/d (rounded).
Strontium (stable)	20	DWEL [8].
	21	Calculated from RfD of 0.6 mg/kg-d [2] (from 1996), assuming 70 kg and 2 L/d (rounded).
Zirconium (stable)	100	No standard benchmark identified; this preliminary indicator was derived from limited toxicity data.
Radionuclides	*Concn (pCi/L)*	*Basis and Context*
Cs-135	*780*	MCL equivalent based on the annual dose of 4 millirem (mrem).
	970	Corresponds to 10^{-4} risk, at 2 L/d over 30 yr.
Cs-137	*110*	MCL equivalent per 4 mrem/yr.
	150	Corresponds to 10^{-4} risk, at 2 L/d over 30 yr.
Sr-90	*36*	Adjusted from 8 pCi/L MCL based on updated dosimetry.
	62	Corresponds to 10^{-4} risk, at 2 L/d over 30 yr.

Target concentrations for the radionuclides incorporate contributions of the radioactive decay products (including short-lived products that would coexist with the parents in the water). These concentrations were developed from (1) MCLs and calculated equivalents (for those without an isotope-specific value), and (2) radiological risk coefficients. The calculated MCL-equivalent values are based on an annual total effective dose equivalent of 4 mrem/yr using the dose conversion factors (DCFs) from FGR 11 [13]. These DCFs reflect biokinetic information for a reference adult male.

The FDA has identified a dose limit for bottled water (in 10 Code of Federal Regulations [CFR] Part 165) that is consistent with the EPA dose-based MCL. That is, the water is "not to contain beta particle and photon radioactivity from manmade radionuclides in excess of that which would produce an annual dose equivalent to the total body or any internal organ of 4 millirems per year calculated on the basis of an intake of 2 L/d. If two or more beta or photon-emitting radionuclides are present, the sum of their annual dose equivalent to the total body or to any internal organ shall not exceed 4 millirems per year." Thus, indicated levels also conform to the FDA limit, which is relevant but not directly applicable to groundwater that may be used as a public drinking water supply. For Cs-135 and Cs-137, the MCL equivalent values are based on the EPA annual dose limit of 4 mrem assuming an ingestion rate of 2 L/d. The 4 mrem/yr limit applies to radionuclides that emit beta particles and photons (X-rays and gamma rays) in drinking water, as indicated above.

Established standards were a primary focus for this evaluation, but where a value is dated and subsequent peer-reviewed toxicity information suggest a different level, further analyses were conducted to assess current knowledge. This is illustrated by the situation for the Sr-90 MCL. That MCL was established many years ago based on the annual dose limit of 4 mrem/yr but using extant dosimetry models. Those earlier dosimetry assumptions have since been updated, including to account for intermediate redistribution, and the concentration based on this more current information is 36 pCi/L. However, the MCL itself has not been revised because the administrative process involved in changing an enforceable standard like an MCL is very protracted and expensive. Thus, absent a pressing regulatory need (e.g., to assure effective health protection), this would not be a priority for the Agency.

For the chemicals, this situation is illustrated by fluoride, for which the MCL is 20 years old. More recent analyses indicate a lower value (by half) may be more appropriate to protect more sensitive subgroups. These further evaluations include the 2001 screening-level review indicated in IRIS [2], documentation of the 2003 minimal risk level [14], and the NAS review of the EPA standards [15]. The lower value recently suggested by the NAS is offered in Table II as an alternate for this chemical.

For the radionuclides, chemical toxicity information was considered in addition to radiotoxicity to assure that leachate targets accounted for both. These chemical toxicity-based levels for stable barium, cesium, strontium, and zirconium are included in Table II. For Sr-90, by comparing target concentrations based on chemical and radiological toxicity, it is clear that the latter represents the limiting value (by a factor of about 300 billion). Similarly, the toxicity of stable Cs-133 is several orders of magnitude lower than that of the radioactive forms.

Many standards are currently being reviewed in light of new information and more recent approaches. To illustrate, EPA has conducted screening-level reviews of the toxicity literature to assess the availability of new information to inform IRIS updates [2]. The Agency is also evaluating more recent information to support the refinement of radiological risk coefficients [16], which are more current than estimators used for current drinking water concentration limits and translate to higher values.

It is important to note that for contaminants naturally present in the environment, target concentrations should be put in the context of ambient levels, including body burdens. For this project, all the study

chemicals are naturally found in the environment, and the radionuclides are ubiquitous as a result of fallout from past atmospheric weapons tests (notably Cs-137 and Sr-90 soil deposition).

SUMMARY AND NEXT STEPS

In an earlier scoping study, three radionuclides and eight chemicals were identified as contaminants of interest for leachate from cesium and strontium capsules stored at the Hanford site. To frame management options for these capsules, it is assumed that contaminants will leach to groundwater and serve as a drinking water source in the long-term future. Before developing performance targets for the initial set of contaminants, a combined fate and toxicity evaluation was conducted to determine if any others should be added to account for decay or fate products and chemical toxicity. From this review, the list was expanded to produce a final study set of 17 contaminants.

Established exposure limits and toxicity data were then reviewed and integrated to develop candidate health-based concentrations to frame performance targets for assessing options for long-term capsule management. This review of more than a dozen different benchmarks and toxicity sources translated to hundreds of individual data checks to support the identification of these target concentrations. In most cases, an EPA drinking water standard served as the key basis for the suggested target. Exceptions were warranted when the drinking water standard reflects dated information and more recent studies suggest a different concentration. Thus, for contaminants with older limits (such as Sr-90), the drinking water standard is identified and an alternate value is also provided that reflects more recent scientific knowledge. Although multiple limits or guidelines exist for most study chemicals, no benchmarks were found for stable cesium or zirconium. Thus, the preliminary target concentrations for these two metals were based on an initial limited review of toxicity data from the scientific literature.

In conclusion, fate and toxicity information can be effectively integrated to develop target concentrations for managing contaminated materials at cleanup sites. These targets are simply intended to serve as guides, not as part of a formal compliance review process. For this reason, to the extent possible, it is useful to incorporate current health-based information relevant to the given application. For the Hanford application, it is expected that the preliminary targets will be refined to reflect evolving knowledge. That knowledge may extend from additional characterization of the capsules and their disposition options, and further context for setting-related conditions, to the refinement of benchmarks and underlying models, and further toxicity assessments. The overall goal is to support technical risk analyses for Hanford with current scientific knowledge and practice, to assure health protection into the long term.

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