DEVELOPMENT OF SITE SPECIFIC RADIOLOGICAL DECOMMISSIONING CRITERIA FOR A HIGH-LEVEL WASTE PROCESSING AND VITRIFICATION PROJECT

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ABSTRACT

In 1980, the United States Congress authorized the U.S. Department of Energy (West Valley Demonstration Project Act; PL-96-368) to solidify 2.1 million litres of liquid high-level radioactive waste (HLW) currently stored at the Western New York Nuclear Service Center (approximately 30 miles south of Buffalo, New York) into a form suitable for transport to and disposal in a federal geologic repository. The method chosen to accomplish this task is vitrification in borosilicate glass.

These wastes were produced during the period 1966-1972 when facilities at the center were used for the only commercial nuclear fuel reprocessing operation in the U.S. These wastes contain approximately 30 million curies of mixed fission products and transuranic radionuclides and are presently being stored in two underground tanks at the West Valley site.

This paper presents the preliminary results of site specific environmental transport and pathway analyses that have been performed to establish radiological decommissioning criteria for final closure of high-level waste processing and treatment facilities. Since neither the derived limits nor the criteria recommended for use as performance objectives have been approved for adoption, this paper serves only to present the methodology used to derive residual contamination criteria for final closure of Project facilities. Decommissioning criteria are expressed in operational units that can be field and laboratory verified (e.g., pCi/gram in soil). These criteria are derived by establishing fundamental performance objectives for public exposure (dose limits) in consort with well defined future intruder and public exposure scenarios.

PURPOSE

An analysis has been performed to estimate the radionuclide concentration limits in soil that would result in an annual committed effective dose equivalent of 100 mrem to a future intruder at West Valley.

Present WVDP policies on soil contamination criteria specify a specific activity of 100 dpm/gram beta plus gamma and 20 dpm/gram alpha (1,2). These criteria were established with the functional objective of "in field" detectability of contamination in excess of background. Although these limits have been useful for operational expediency they are, in fact, within the general range and variability of soil background in the U.S. (3) for naturally occurring radionuclides (potassium-40, uranium, radium, etc.).

Therefore, the analysis described herein was undertaken from a dosimetric/risk perspective. A secondary objective was to develop limits that could be used to provide relief from the need to store and/or dispose of large volumes of relatively clean soil which, if considered nonradioactive with no restrictions placed on disposition, would pose a minimum and acceptable risk to future inhabitants of the site. These same limits will be applicable as residual contamination limits for final closure of Project facilities.

The 100 mrem/yr exposure criterion has been previously recommended as a performance objective for decontamination and decommissioning prior to final closure of the WVDP (4,5,6). These documents provide justification for this value and contain examples of the application of this value in establishing acceptable long term residual radiological risks from nuclear facilities under various exposure scenarios including the resident agricultural intruder described here.

Since neither the derived limits nor the criteria recommended for use as performance objectives have been approved for adoption, this paper serves only to present the methodology used to derive residual contamination criteria for final closure of Project facilities.

A residential farming scenario, in which a family constructs a home at on-site locations and raises an appreciable fraction of their food is considered to be a credible bounding case for performance assessment (7). Important parameters which, in general, define the resident farmer scenario have been previously identified (7,8,9,10,11) and are discussed below. It has been assumed that homesteading (initiation of exposure) begins immediately after a 100 year institutional control period following final closure of the WVDP. That is, exposure begins about the year 2100

AD. However, the recommended soil limits have been derived for application during the active Project period prior to initiation of institutional control.

GENERAL DESCRIPTION OF INTRUDER SCENARIO

For purposes of our analysis, we assume that contaminated soil has been dispersed across and into the ground surface with no regard for the radiological character of the soil, i.e., the site is covered to an effectively infinite areal extent and depth. In fact, a surface area of ≥ 1 ha and a depth of ≥ 1 metre can be considered as "effectively infinite" (see Ref. 7). An "effectively-infinite slab" source at the surface of the earth is therefore assumed. The near surface gravel/sand regime of the north plateau area is the only credible on-site area at the WVDP from which a future intruder could extract sufficient quantities of water to sustain his family and farm. Therefore, this location is assumed to provide a source of contaminated water for human and livestock consumption.

A farmer and his family were assumed to reside throughout their lifetimes on this site, produce all their water, milk, vegetables, and meat from their farm which is located entirely within this "contaminated" soil regime. We therefore assume the following exposure pathways are relevant:

- external exposure to contaminated soil
- inhalation of suspended activity from the soil
- ingestion of contaminated drinking water
- ingestion of milk and meat from the dairy and beef cattle that drink contaminated water
- ingestion of vegetables grown in the contaminated soil
- ingestion of contaminated soil from the vegetable garden

GENERAL METHODOLOGY AND DEFINITION OF PATHWAY MODELS

The general form for the soil concentration limit can be simply expressed as:

$$C = \frac{D_o}{D/S} \qquad (1)$$

where:

C = Radioactivity concentration in soil (e.g., pCi/gram)

D_o = Dose objective (e.g., 100 mrem/yr)

D/S = Dose to source ratio for a unit concentration of radioactivity in the soil (e.g., mrem/yr per pCi/gram)

The assumed nuclide mixture in soil ("source term") is the former reprocessing plant spent fuel distribution normalized to unit concentration of Cs-137 as defined in Ref. 12. Therefore, D/S can be expressed in mrem/yr per unit concentration (pCi/gram) cesium-137 soil contamination provided that the dose contribution from all important nuclides associated with a unit concentration of cesium-137 have been considered in the analysis. Since homesteading is not assumed to occur until 100 years following WVDP closure (5), the 1987 nuclide distribution must be decay corrected to represent the distribution and radiological conditions of exposure in the year ≈ 2100 AD.

However, the derived limits can then be "reverse decay" corrected to represent concentration limits during the active phase of the Project (present time to 2000 AD). The relevant nuclide mixes for the time periods of interest are shown in Table I. The dose/source ratio is evaluated as follows:

$$D/S = \sum_{ip} (C_{ip})(U_{ip})(DCF_{ip}) \qquad (2)$$

where:

D/S = Dose/source ratio expressed as the committed effective dose equivalent (50 years) per unit concentration cesium-137 (e.g., mrem/yr per pCi/gram cesium-137 in soil)

C_{ip} = Concentration of nuclide i in medium of exposure for pathway p (air, water, soil, food stuffs)

U_{ip} = Usage parameter; annual exposure time or annual intake for nuclide i and pathway p.

DCF_{ip} = Dose Conversion Factor; committed effective dose equivalent per unit concentration or intake of nuclide i in pathway p.

By setting the concentration of cesium-137 in soil at unity (1 pCi/gram), the resultant contribution to the D/S ratio can be evaluated for each nuclide (in accordance with its concentration normalized to cesium-137, see Table I) and for each exposure pathway relevant to the resident farmer scenario. Equation 1 can then be evaluated once the total D/S ratio is derived. The general models and equations for each exposure pathway are presented below.

TABLE I

Major Components of the Source Term (Radioactive Mix)-West Valley Spent Fuel Distribution.

Nuclide	Activity Normalized to Cs 137-Year 1987	Half Life (Yrs)	Activity Normalized to Cs 137-Year 2100
Sr/Y 90	0.94 E+0	28	0.79
Cs 134	1.9 E-3	2.1	~0
Cs 137/Ba 137m(1)	1.0 E+0	30	1.0
Pm 147	4.2 E-2	2.6	~0
Eu 154	1.8 E-2	16	~0
Pu 238	4.9 E-2	86	0.27
Pu 239	1.3 E-2	$>10^4$	0.18
Pu 240	9.9 E-3	$>10^3$	0.14
Pu 241	6.6 E-1	13.2	~0
Am 241(2)	3.7 E-2	460	0.73
Am 243	3.3 E-4	$>10^3$	4.5 E-3
Cm 244	3.0 E-3	17.6	~0

(1) Barium 137m/cesium-137 ratio actually equal to 0.94; taken to be 1.0 for analysis purposes.

(2) Americium 241 ingrowth from Pu 241 taken as theoretical maximum of 0.025 of 1987 Pu 241 activity in Year 2100.

DEFINITION OF THE DOSE/SOURCE RATIO FOR EACH SPECIFIC EXPOSURE PATHWAY

In this section, Eq. (2) is evaluated for each exposure pathway to derive pathway specific expressions for the resident farmer scenario. The following subsections present the model equations for estimating the D/S ratio for each exposure pathway.

External Exposure to Contaminated Soil

We have assumed a uniform and homogeneous concentration of radionuclides in an effectively infinite soil regime. However, two exposure conditions are defined for this pathway: 1) The intruders spend some fraction of time per year working in the vegetable gardens that produce 100 percent of their vegetables and are therefore exposed directly to photons emitted by the contaminated soil; and 2) the intruders spend an appreciable portion of their time indoors in their home which affords some shielding from the contaminated soil surrounding and under the house. Equation 3 below is evaluated separately for each condition and the results summed. For the external exposure pathway, the D/S is evaluated as follows:

$$D_{ie}/S = (C_{is})(U_e)(SF)(DCF_{ei})(\rho_s) \qquad (3)$$

where:

D_{ie}/S = Dose to source ratio for nuclide i for the external exposure pathway (mrem/yr per pCi/gram cesium-137)

C_{is} = Concentration of nuclide i in soil (pCi/gram)

U_e = Usage factors for the external exposure pathway i.e., fraction of time spent in the garden out of doors or inside the home (unitless)

SF = Shielding factor if applicable (unitless)

DCF_{ei} = Dose conversion factor for the external exposure pathway, nuclide i (mrem/yr per pCi/cm^3)

ρ_S = density of soil (gram/cm^3)

Inhalation of Suspended Activity from the Soil

As discussed previously, although we have assumed a uniform distribution of activity throughout an effectively infinite soil regime, two distinct exposure conditions are assumed: 1) out of doors working in the vegetable garden; and 2) inside the house. Each condition is evaluated separately and the results summed. We have estimated the concentration of suspended material (dust) in air using a mass loading approach (13) which is based on observations of airborne concentrations of naturally occurring materials, such as uranium and thorium, relative to concentrations in surface soils. For the inhalation (airborne) pathway, the dose/source ratio is evaluated as follows:

$$Dia/S = (C_{is})(M_a)(U_a)(F)(BR)(DCF_{ia}) \qquad (4)$$

where:

Dia/S = Dose to source ratio for the airborne pathway, nuclide i (mrem/yr per pCi/gram cesium-137)

M_a = Mass loading factor of dust in air under stated conditions (g/m^3)

U_a = Usage factor for the inhalation pathway for outdoors in the garden or indoors in the house (unitless)

F = Filtration factor, if applicable, i.e., ratio of indoor dust concentration to outdoor dust concentration (unitless)

BR = Breathing rate (m^3/yr)

DCF_{ia} = Dose conversion factor for the inhalation pathway, nuclide i (mrem per pCi annual intake)

Ingestion of Contaminated Drinking Water

Critical to the evaluation of the D/S ratio for the water related pathways are assumptions regarding the transfer of radionuclides from the soil regime to water percolating or flowing through it. The distribution factor (Kd) approach has been used in this analysis for this purpose using representative values from the literature. This factor relates the concentration in soil to that in water in contact with it, expressed as the ratio of activity concentration in soil per activity concentration in water and has the units of mL/g. Admittedly, analysis is quite sensitive to the choice of Kd and unfortunately a wide range of values are reported in the literature for each nuclide of interest. Reported values are typically geologically and geochemically specific to a stated circumstance, environment, and/or site.

Values believed to be representative for the sand/gravel regime and local geochemical conditions at West Valley were selected. However, Kd's are quite site specific and for future assessments of this type, site specific data (e.g., bench/column tests) should be obtained.

The concentration of nuclide i in water extracted from the north plateau sand/gravel regime and used for drinking water and livestock is defined simply as:

$$C_{iw} = \frac{C_{is}}{Kd_i} \tag{5}$$

where:

C_{iw} = Concentration of nuclide i in water used for drinking and livestock (pCi/mL)

Kd_i = Distribution coefficient for nuclide i (mL/gram)

The drinking water pathway is therefore modeled as follows:

$$D_{iw}/S = (C_{iw})(I_w)(DCF_{ii}) \tag{6}$$

where:

D_{iw}/S = Dose to source ratio for the drinking water pathway, nuclide i (mrem/yr per pCi/gram cesium-137)

I_w = Annual intake of water by intruder (mL)

DCF_{ii} = Dose conversion factor for the ingestion pathway, nuclide i (mrem per pCi annual intake)

Ingestion of Milk and Meat

It is assumed that 100 percent of the milk and meat consumed by the intruder is produced at their farm from their dairy and beef cattle. The cattle obtain all drinking water from the same source the intruders use for drinking water (the sand/gravel regime of the north plateau). The contributions to the dose/source ratio from consumption of contaminated milk (m) and meat (b) are modeled as follows:

$$D_{im}/S = (C_{iw})(Q_m)(F_{im})(I_m)(DCF_{ii}) \tag{7}$$

$$D_{ib}/S = (C_{iw})(Q_b)(F_{ib})(I_b)(DCF_{ii}) \tag{8}$$

where:

D_{im}/S and D_{ib}/S = Dose to source ratios for the milk and meat ingestion pathways respectively, nuclide i (mrem/yr per pCi/gram Cs-137)

Q_m and Q_b = Daily intake of water by dairy cows and beef cattle respectively (mL/day)

F_{im} and F_{ib} = Equilibrium ratios for dairy cows and beef cattle respectively; concentration in milk or meat relative to daily intake (pCi/mL in milk or pCi/g in beef per pCi/day intake)

I_m and I_b = Annual consumption by intruder of milk (mL) and beef (g) respectively

Ingestion of Vegetables

It is assumed that 100 percent of the intruders' vegetable consumption comes from their vegetable gardens. Therefore, all vegetables in the intruder's diet are grown in contaminated soil. In our model we have not made distinctions between vegetable types and have used general soil to plant transfer coefficients (uptake factors which are, however, chemical element specific) from the literature. We have also assumed that the radionuclide inventory available to the plants is taken up exclusively by the roots from the soil. We believe that additional radionuclide uptake through irrigation of food crops is not valid for this scenario in the humid, high precipitation environment of West Valley. The vegetable ingestion pathway is therefore modeled as follows:

$$D_{iv}/S = (PS_{iv})(C_{is})(I_v)(DCF_{ii}) \tag{9}$$

where:

D_{iv}/S = Dose to source ratio for the vegetable ingestion pathway, nuclide i (mrem/yr per pCi/gram cesium-137)

PS_{iv} = Soil to plant transfer coefficient, nuclide i, (pCi/gram wet weight in vegetable per pCi/gram dryweight in soil)

I_v = Annual consumption of vegetables by intruder(grams/yr)

Ingestion of Contaminated Soil from the Vegetable Garden

It is assumed that while working in the vegetable gardens (transfer of soil from hands to mouth) and as a result of inadequate washing of vegetables prior to eating them, the intruders routinely ingest small quantities of contaminated soil directly from the gardens. For the soil ingestion pathway, the dose/source ratio is evaluated as follows:

$$D_{is}/S = (C_{is})(I_s)(DCF_{ii}) \qquad (10)$$

where:

D_{is}/S = dose to source ratio for the soil ingestion pathway, nuclide i (mrem/yr per pCi/gram cesium-137)

I_s = rate of soil ingestion (grams/yr)

EVALUATION OF PATHWAY SPECIFIC EXPRESSIONS

Dose Calculations

This section presents the data used in estimating the dose to source ratios (annual committed effective dose equivalent per unit concentration in soil) for each of the pathways of exposure that define our resident agricultural intruder scenario. Each pathway expression Eq. (3-10) is evaluated for each important nuclide (see Table I) and the results summed to provide the D/S ratio for each pathway. The resultant D/S ratios are then summed to arrive at a total D/S ratio for the resident agricultural intruder scenario. Equation 1 is then evaluated. These results are presented in Table II.

The dose analysis assumes that the probability of occurrence for each exposure pathway is unity and that intrusion can occur at any time after the institutional control period of 100 years after WVDP closure. Since these are conservative assumptions which tend to overestimate the risk to intruders, we have attempted to choose reasonable average values for parameters that describe transport of radionuclides through food chains, for annual intakes and exposure times rather than maximum possible values (14). This approach is considered consistent with the intent of the International Commission on Radiological Protection (ICRP) that limits on radiation dose should apply to average individuals within a critical group of maximally exposed individuals rather than the single, theoretical individual who might receive the highest possible dose (15).

Determination of Soil Concentration Limits for the WVDP

The total dose to source ratio [D/S in Eq. (1)] is therefore a simple sum of dose to source ratios determined for each individual exposure pathway and nuclide and is expressed as follows. All units are mrem/yr per pCi/gram Cs-137 in soil.

TABLE II

Dose to Source Ratio for the Residential Farmer Intruder Scenario.

Nuclide	External Exposure	Inhalation in Garden	Inhalation Indoors	Water Ingestion	Milk Ingestion	Beef Ingestion	Vegetable Ingestion	Soil Ingestion
Sr 90/Y 90	~0	4.3 E-4	9.6 E-5	2.0	5.1 E-2	7.5 E-3	1.5	4.0 E-3
Cs 137/Ba 137 m	3.9	1.4 E-5	3.2 E-6	1.8 E-1	2.3 E-2	4.5 E-2	4.3 E-2	1.8 E-3
Pu 238	~0	5.5 E-1	1.2 E-2	2.2 E-2	4.0 E-8	2.7 E-7	7.8 E-4	3.9 E-3
Pu 239	~0	4.1 E-1	9.4 E-3	1.6 E-2	2.7 E-8	1.8 E-7	5.2 E-4	2.9 E-3
Pu 240	~0	3.2 E-1	7.3 E-3	1.2 E-2	2.0 E-8	1.4 E-7	4.0 E-4	2.2 E-3
Am 241	~0	1.7	3.9 E-2	1.2 E-1	9.0 E-7	5.5 E-6	3.2 E-3	5.9 E-2
Am 243	~0	1.1 E-3	2.4 E-4	7.6 E-4	6.0 E-9	4.0 E-8	2.0 E-5	3.6 E-4
Subtotals	3.9	3.0	6.8 E-2	2.3	7.4 E-2	5.3 E-2	1.6	7.4 E-2

Total for all pathways: 11.1 mrem/yr per pCi/gram cesium-137

$$D/S = D_e/S + D_a/S + D_w/S + D_m/S + D_b/S + D_v/S + D_s/S$$

With all terms as previously defined. This summation is evaluated and is presented in Table II. The overall D/S for the resident agricultural intruder scenario at WVDP is therefore calculated to be:

D/S = 11 mrem/yr per pCi/gram cesium-137 in soil

Equation 1 can now be evaluated since D/S is known:

$$C = \frac{D_o}{D/S} = \frac{100 \text{ mrem/yr}}{11 \text{ mrem/yr per pCi/gram}} = 9.1 \text{ pCi/gram Cs-137}$$

This Cs-137 concentration (and associated nuclides at the Table I normalized concentrations for the year 2100 AD) represents the radiological conditions of exposure from which our future intruder will receive a dose ≤100 mrem/yr.

For times of exposure much beyond our assumed 100 year institutional control period (beyond ≈2100 AD), this concentration limit will actually increase due to radiological decay. (Additionally, we have taken no credit for "environmental turnover" of the soil during the 100 year institutional control period which is a highly conservative assumption.) Therefore, the value of D/S = 9.1 calculated for the year ≈ 2100 AD represents a "maximum" value (during the intruder's life time) within the envelope of assumptions inherent in the analysis.

The concentration limit (9.1 pCi/gram) must now be "reverse decayed" to be representative of the cesium-137 concentration at the present time (1987 to≈2000 AD) that would ultimately result in a 100 mrem/yr exposure 100 years later. This is done simply using the fundamental law of radioactive decay.

Since the half life of Cs-137 is 30 years, the "present time frame" radiological concentration limit which meet the dose objective (100 mrem/yr) established for the future time frame (τ = 100 yrs) is therefore estimated to be 91 pCi/gram Cs-137.

This value can also be expressed as gross alpha and/or gross beta/gamma activity by revisiting the details of the assumed present time frame (1987) spent fuel nuclide distribution. The following nuclides from Table I are considered "alpha emitters" for purpose of this analysis: Pu 238-240, Am 241/243, Cm 244. All other nuclides listed in Table I are considered "beta/gamma" emitters.

Summing the appropriate normalized activities of Table I (1987 column) results in an alpha/Cs-137 ratio of 0.113 and a beta/gamma ratio relative to Cs-137 of 4.54.

Regarding beta/gamma emitters, it is assumed that the Ba-137m/Cs-137 ratio is 0.94 and that the Sr-90 and Y-90 total activity ratio relative to Cs 137 is 1.9 (2 x 0.95). It should also be noted that this analysis has ignored the specifics of nuclear decay schemes and relative yields in identifying nuclides as "pure alpha emitters." Additionally, the relatively small concentrations of uranium isotopes (activity basis) have been ignored since consideration will not impact the results.

The associated "gross alpha" and "gross beta/gamma" activities can then be calculated:

$$C_\alpha = (91 \text{ pCi/g Cs-137}) \left(\frac{0.113 \text{ pCi } \alpha}{\text{pCi Cs-137}}\right) = 10 \text{ pCi/g} = 22 \text{ dpm/g}$$

$$C_{\beta/\gamma} = (91 \text{ pCi/g Cs-137}) \left(\frac{4.54 \text{ pCi } \beta+\gamma}{\text{pCi Cs-137}}\right) = 410 \text{ pCi/g} = 920 \text{ dpm/g}$$

Comparison with the ONSITE/MAXI Computer Code

Since it was recognized that the intruder impacts calculated for the contaminated soil case above are similar to those calculated for on-site disposal of radioactive waste under 10 CFR 20.302, the ONSITE/MAXI1 computer code (13) was run to provide an independent check on the calculations.

As with the model utilized in the earlier sections, irrigation of food crops is not assumed to occur in the humid, high precipitation environment of West Valley. There are, however, some small differences in the ONSITE/MAXI1 model. For example, the feed for the cattle is assumed to be grown on-site which tends to increase the radionuclide concentrations in the meat and milk. Other differences include the model used for external exposure, the use of ten different food types, holdup times, translocation factors, food consumption rates, and specific values for various parameters in Eq. (1-10).

The results obtained using the ONSITE/MAXI1 model agree favorably (within a factor of 3) with the results obtained in the rigorous analysis in the preceding subsections. Therefore, the values calculated manually using Eq. (1-10) are judged to be valid and are thus recommended for operational adoption.

CONCLUSIONS

It was shown that for a unit concentration of the assumed nuclide mix (source term), the dose to source ratio (D/S) for our future agricultural intruder is estimated to be

11 mrem/yr per pCi/gram Cs-137. This ratio is representative of the condition of exposure 100 years after WVDP closure. Using a dose limitation for the future intruder of 100 mrem/yr results in a recommended soil limit for present application (during WVDP active Project phases) of 91 pCi/gram cesium-137. Since neither the derived limits nor the criteria recommended for use as a performance objective have been approved for adoption, this paper serves only to present the methodology used to derive residual contamination criteria for final closure of Project facilities. Some useful insights into the components of the hypothetical intruder dose include:

- < 15 percent of the dose results from plutonium and americium isotopes (all pathways);
- Approximately 30 percent of the dose results from external exposure to Cs-137/Ba-137m; and
- Approximately 50 percent of the dose results from ingestion of Sr-90/ Y-90

The dose to source ratio is dominated (80 percent) by the beta/gamma emitters. Therefore, assuming a nuclide mix generally similar to the one we have analyzed regarding beta/gamma to alpha emitter relationships (Sr/Y + Cs/Ba vs Pu/Am), the recommended gross beta/gamma limit of ≈ 900 dpm/gram should be considered controlling. Gross alpha contamination levels several times higher than the derived limit (22 dpm/gram) will not significantly increase risks from exposure, provided that the gross beta/gamma limit is clearly achieved. However, for nuclide mixes which contain significantly higher Pu and/or Am components than those assumed here, this analysis should be revisited to establish criteria appropriate for that nuclide mix.

The ONSITE/MAXI1 Code (13) also was used to determine the dose to source ratio from all relevant pathways associated with a unit concentration of the assumed nuclide mix. It was shown that similar results are obtained relative to the WVDP model presented here.

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