

ASSESSMENT OF RISK FROM LAND APPLICATION OF WASTE WATER CONTAINING ELEVATED LEVELS OF NATURALLY OCCURRING RADIOACTIVE MATERIALS¹

by

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Abstract. On-site land application of waste water containing very low levels of site-derived contaminants is a commonly used method of disposal. However, the residual soil contamination resulting from such practices may complicate and increase the costs of site remediation if constituent soil concentrations exceed regulatory clean-up standards. The long-term potential human health impacts of land application of waste water containing elevated levels of naturally occurring radionuclides for a hypothetical in-situ uranium facility were investigated using the U. S. Department of Energy RESRAD Computer Code for assessment of dose and risk. The post-reclamation radiation doses in excess of background, attributable to land application of waste water containing uranium and its decay products thorium and radium, were calculated for several different conditions assuming rural-residential use of the land. For contaminated land areas of 10 acres or more, the estimated doses were directly proportional to the soil radionuclide concentration when the thickness of the contaminated soil and hydrological parameters were held constant. The maximum annual radiation dose to a rural resident on land with surface soil concentration equal to 205 pCi/g natural uranium was estimated to be 34 mrem. In contrast, the maximum annual radiation dose to this hypothetical individual from ²²⁶Ra at the residual concentration allowed under the regulations for uranium mill tailings, i.e., 5 pCi/g, varied from 135 mrem to 283 mrem depending on the thickness of the contaminated layer.

Additional Key Words: RESRAD, dose, exposure pathways, reclamation, uranium.

Introduction

On-site land application of large volumes of waste water containing elevated levels of natural uranium and its decay products (i.e., radium-226 [²²⁶Ra] and thorium-230 [²³⁰Th]) can be an efficient and environmentally sound method of disposal of waste water. In cases where the concentrations of naturally occurring radionuclides in waste water are elevated only slightly above regional background values, the concentrations in soil attributable to the waste water disposal may be indistinguishable from soil background concentrations. However, where waste water radionuclide concentrations are significantly elevated, the long-term risk to human health and the potential for exceeding regulatory standards

for site reclamation must be considered prior to the implementation of a waste water land application program.

Regulatory Requirements for Reclamation of Contaminated Sites

The U. S. Environmental Protection Agency (EPA) has proposed a radiation dose criterion for clean-up of contaminated sites, other than uranium mill tailings disposal facilities, of 15 mrem per year, total effective dose equivalent (TEDE) from all exposure pathways. The Nuclear Regulatory Commission (NRC) has proposed a similar rule. (As of March 15, 1997, neither of these proposed rules has been finalized and the dose criteria that will be included in the final rules are still in question.)

Reclamation of uranium mill tailings is a special case, regulated by the NRC under the Code of Federal Regulations Section 10 Part 40 (10 CFR 40) and by "agreement states" under compatible regulations. These regulations require uranium mill tailings to be cleaned up to a specific radionuclide concentration in soil rather than a projected human dose limit, i.e. 5 pCi ²²⁶Ra per gram (5 pCi/g) above background in the top 15 cm of soil and 15

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pCi ^{226}Ra per gram (15 pCi/g) for soils at depths greater than 15 cm.

The activity concentration limit for ^{230}Th is based on limiting the total ^{226}Ra concentration over a period of 1,000 years. Thorium decays to ^{226}Ra . After 1,000 years, the ^{226}Ra activity concentration due to decay of thorium will reach approximately 35 percent of its equilibrium value, or 35 percent of the activity concentration of the ^{230}Th . At this time there is no specific regulatory limit for uranium in soil for uranium mills.

RESRAD Dose Assessment Code

Since low level radiation doses cannot be directly measured, demonstrating compliance with the regulatory criteria requires the use of radiation dose modeling. For regulatory purposes, the NRC accepts the results of modeling using the RESRAD computer code developed by the U.S. Department of Energy (DOE) as a tool for implementing residual radioactive material guidelines (Yu, et al., 1993).

RESRAD calculates doses to individuals from all exposure pathways based on the assumed land use (Figure 1). Radionuclide contamination in soil can expose human receptors directly through ingestion of soil, resuspension as inhalable dust, emanation of radioactive gas, i.e., radon, and direct gamma radiation or x-rays. Indirect exposure pathways include ingestion of ground water containing elevated radionuclide levels, ingestion of foods grown on contaminated soil or

irrigated with contaminated water, and ingestion of fish from surface water impacted by soil contamination at the site. RESRAD can be used to estimate potential dose from exposure to multiple radionuclides and identify critical exposure pathways and to project doses into the future.

Methods

The potential doses to residents on land that had been previously used for land application of waste water containing elevated levels of uranium and its decay products were estimated for time periods up to 1,000 years post-reclamation. Waste water concentrations and volumes were based on a hypothetical facility; however, the values used are representative of real situations. The analysis was principally designed to evaluate the effect on potential risk of uranium contamination in soil at various concentrations and under differing conditions with regard to extent of the contaminated surface area, specific soil characteristics, annual precipitation rates, and depth to ground water.

Radionuclide Concentration in Soil

Two scenarios were used with regard to uranium concentration in soil; Case 1, involving constant total activity, and Case 2, using a constant activity concentration. For Case 1, the activity of uranium in waste water, applied to the land, was kept constant while other parameter values, such as thickness and area of the contaminated zone, were varied. For Case 2, the excess natural uranium concentration was maintained at a constant level, 45 pCi/g.

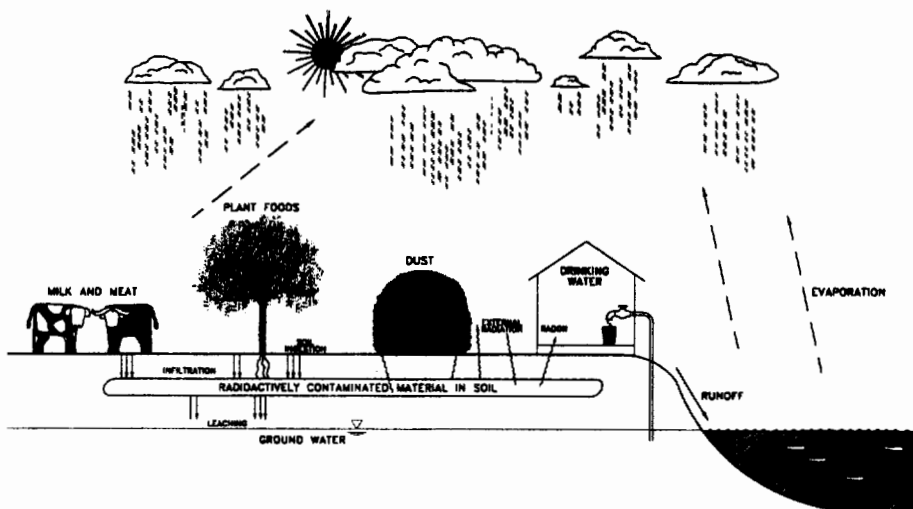


Figure 1. Exposure Pathways Considered in RESRAD

For Case 1, the initial uranium concentrations in soil were calculated in the following manner:

Area of the land application unit: 20 acres
($8.09 \times 10^4 \text{ m}^2$)

Annual volume of waste water applied: 100 acre-ft
($1.23 \times 10^8 \text{ L}$)

Tilling depth: 0.15 m (NCRP 1996)

Land application duration: 15 years

Uranium concentrations in waste water: 3 mg/L
(3 ppm)

Assuming the waste water was uniformly applied over a 20 acre field, the concentration of uranium in soil attributable to the disposal process would be as follows:

$$M_s = AD\rho CF \quad (1)$$

where: M_s = mass of affected soil (g)
A = area of land application (m^2)
D = tilling depth (m)
 ρ = density of soil (g/cm^3)
CF = conversion factor ($10^6 \text{ cm}^3/\text{m}^3$)

$$M_s = (8.09 \times 10^4 \text{ m}^2)(0.15 \text{ m})(10^6 \text{ cm}^3/\text{m}^3)(1.5 \text{ g}/\text{cm}^3) \\ = 1.82 \times 10^{10} \text{ g}$$

The total mass of uranium applied to the soil over a 15 year period would be as follows:

$$M_u = C_u V \quad (2)$$

where: M_u = mass of uranium added to soil (mg)
 C_u = concentration of uranium in waste water (mg/L)
V = volume of waste water applied to the land

$$M_u = (3 \text{ mg}/\text{L})(1.23 \times 10^8 \text{ L}/\text{y})(15 \text{ yr}) \\ = 5.55 \times 10^9 \text{ mg}$$

Assuming that ^{234}U and ^{238}U are in equilibrium and that the specific activity of ^{238}U is 330 pCi/mg, the activity concentrations of the two isotopes attributable to the land application of waste water would be as follows:

$$C_{us} = M_u SA_u / M_s \quad (3)$$

where: C_{us} = activity concentration of uranium in soil
 SA_u = specific activity of uranium

$$C_{U-238\text{-soil}} = (330 \text{ pCi}/\text{mg})(5.55 \times 10^9 \text{ mg}) / (1.82 \times 10^{10} \text{ g soil}) = 100 \text{ pCi}/\text{g}$$

At equilibrium, the concentration of ^{234}U in soil would be equal to the ^{238}U concentration or 100 pCi/g. The activity of ^{235}U in natural uranium is approximately equal to 4.5% of the ^{238}U activity or 4.5 pCi/g. The natural uranium (U-nat) activity concentration, i.e., the sum of the activity concentrations for the three uranium isotopes, would be 205 pCi/g.

For the Case 1 analysis, the total activity of uranium applied to the land was maintained at a constant value:

^{238}U	1.8 Ci (1.8×10^{12} pCi)
^{235}U	0.081 Ci (8.1×10^{10} pCi)
^{234}U	1.8 Ci (1.8×10^{12} pCi)
U-nat	3.7 Ci (3.7×10^{12} pCi)

For the Case 2 analysis, the total activity of uranium applied to the land varied but the concentration remained constant with the total excess natural uranium (U-nat) inventory ranging from 0.82 Ci (8.2×10^{11} pCi) to 4.1 Ci (4.1×10^{12} pCi).

For this case, the excess natural uranium concentration in soil was maintained at a constant level of 45 pCi/g, based on a reasonable hypothetical situation. The area of the contaminated zone was varied from 20-100 acres.

The RESRAD analysis was also performed assuming the soil concentrations of ^{226}Ra to be at the clean-up standard for uranium mill tailings, i.e., 5.0 pCi/g above background. The ^{230}Th concentration in soil was also assumed to be at 5.0 pCi/g as this is the de facto standard for soils with ^{226}Ra concentration at the 5.0 pCi/g limit.

RESRAD Analysis

Doses were estimated using RESRAD and default dose conversion factors, food transfer factors, bioaccumulation factors, and distribution coefficients for specific radionuclides, including the short-lived decay products. Exposure pathways and lifestyle parameter values for rural residential land use were employed throughout the analysis. Parameter values that were varied in the analysis included thickness of the contaminated zone, soil type (hydraulic conductivity and porosity), depth to ground water, and area of the contaminated zone.

Doses were estimated for both soil concentrations with user selected parameter values for area of contamination, thickness of contaminated layer, annual precipitation rate, depth to ground water, and soil type. Default values were used for other parameters.

Using RESRAD, annual dose rates were projected for time periods extending from 0 to 1,000 years after the soil was contaminated. The maximum time period of 1,000 years was selected since regulations regarding uranium mill tailings specify control for 1,000 years. For the purpose of this analysis, exposure was assumed to start the year following cessation of land application of waste water.

Results

The results of the RESRAD analyses for Case 1 are given in Tables 1 and 2. User defined parameter values for each set of analyses are specified as footnotes to the Tables. Default values were used for other RESRAD input parameters. Table 1 shows the projected annual dose rates by pathway. Table 2 gives the projected annual total dose rate for all pathways over time.

For Case 1, where the total activity applied to the land was held constant, the soil concentration was inversely proportional to the thickness of the contaminated layer at time 0 (Table 1). Based on the RESRAD analysis, the estimated doses were also nearly inversely proportional to the thickness of the contaminated zone, i.e., proportional to the concentration. However, there was a trend towards higher doses per unit concentration as the thickness of the contaminated layer increased from 0.15 to 1.0 meter due primarily to increased plant uptake as uranium reached the root zone. The total dose per pCi/g uranium, at time 0, ranged from 0.17 mrem/year for a 0.15 meter thick contaminated zone to 0.25 mrem/year for a 2 meter thick contaminated zone.

When the uranium concentration in soil and the depth of the contaminated zone was held constant and the area of the contaminated zone was varied (Case 2), the estimated annual dose, 8 mrem/year, was independent of the area. The projected annual dose rate per pCi/g was 0.17 mrem/year. The range of contaminated zone areas used in this investigation was limited to areas which might actually be used in a land application program. Within this range, the area of the contaminated zone does not affect the estimated dose for the year immediately after the contamination occurred (time 0), when the concentration is kept constant (Case 2).

For both cases, as would be expected, the projected annual doses during the first 30 years are dominated by surface effects, primarily inhalation of resuspended particulates and direct gamma radiation exposure. The projected annual doses from ground water pathways 100 years or more after reclamation, were dependent on the parameter values selected for soil characteristics and depth to ground water.

In contrast to the estimated doses from uranium contamination in soil, the analysis for a constant ^{226}Ra soil concentration of 5 pCi/g shows a significant increase in dose with increased thickness of the contaminated zone. The rural residential land use in RESRAD assumes that a residence is built directly on or over the contaminated zone. Therefore, as would be expected and as shown by the analyses, the principal contributor to the dose is radon-222, a radioactive gas formed in the soil by the decay of ^{226}Ra which can infiltrate into the house. The estimated annual dose per pCi/g ^{226}Ra was 26.9 mrem/year for a 0.15 meter thick contaminated zone to 56.6 mrem/year for a 2.0 meter thick contaminated zone. The estimated annual dose per pCi/g U-nat ranged from 0.17 mrem/year for a 0.15 meter thick contaminated zone to 0.25 mrem/y for a 2.0 meter thick contaminated zone.

Conclusions

The estimated doses which might be incurred by future residents on land used for disposal of waste waters containing elevated concentrations of naturally occurring radionuclides may exceed regulatory criteria for clean-up of contaminated sites. The potential for creating a situation where resources may need to be used to clean-up such sites should be investigated using a dose assessment code such as RESRAD prior to implementation of the waste water disposal process. Site specific parameter values such as depth to ground water, soil type, and annual precipitation rate are critical to the analysis.

The land application waste disposal program should be designed to balance current costs against potential soil clean-up costs. While it is difficult to predict the direction regulatory requirements will take in the future, a conservative approach taken in regard to land application may limit or prevent clean-up costs in the future.

Table 1. Variation in Estimated Annual Dose (mrem/year) by Pathway with Thickness of Contaminated Layer (Excess Uranium Inventory Constant - Case 1)

Thickness	0.15 m	1.0 m	2.0 m
U-nat Concentration (pCi/g)	205	30.8	15.4
	Estimated		
Pathway	Annual Dose (mrem/year)		
Ground	8.91	1.66	0.89
Inhalation	19.16	2.88	1.54
Plant	2.08	1.86	0.99
Meat	0.64	0.12	0.07
Milk	1.64	0.30	0.18
Soil	1.55	0.23	0.12
Water	0.00	0.00	0.00
TOTAL	33.98	7.05	3.79
Annual Dose per pCi/g	0.17	0.23	0.25

¹ Soil Type - Clay

² Depth to Ground Water - 100 m

³ Annual Precipitation Rate - 0.5 m

⁴ Area of Contamination - 20 acres ($8.09 \times 10^4 \text{ m}^2$)

Table 2. Variation in Total Estimated Annual Dose (mrem/year) with Thickness of Contaminated Layer (Excess Uranium Inventory Constant - Case 1)

Thickness	0.15 m	1.0 m	2.0 m
U-nat Concentration (pCi/g)	205	30.8	15.4
Years After Reclamation	Total Estimated Annual Dose (mrem/year)		
0	33.99	7.08	3.75
1	32.71	7.04	3.74
3	30.27	6.98	3.73
10	23.07	6.73	3.66
30	10.50	6.11	3.49
100	0.51	4.35	2.96
300	0.00	1.59	1.87
1000	0.00	0.00	0.00

¹ Soil Type - Clay

² Depth to Ground Water - 100 m

³ Annual Precipitation Rate - 0.5 m

⁴ Area of Contamination - 20 acres ($8.09 \times 10^4 \text{ m}^2$)

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Appendix 1: Glossary

Curie: The curie (Ci) is basic unit used to describe the amount of radioactivity present. One curie of a radioactive material will undergo 37 billion (3.7×10^{10}) disintegrations per second and is approximately equal to the amount of radioactivity in one gram of ²²⁶Ra.

Picocurie: A picocurie (pCi) is the activity unit commonly used in relation to soil contamination and is equal to 10-12 curies.

Rem: The unit of radiation dose commonly used in radiation protection is the rem. The rem takes into account not only the amount of energy absorbed by the body but also the energy distribution and, when used to express effective dose, includes a weighting factor to account for the sensitivity of the organs or tissues receiving the dose.

Millirem: The unit of dose generally used in relation to clean-up standards is the millirem (mrem).

TEDE: The Total Effective Dose Equivalent or TEDE is the total effective dose to the body including dose from external radiation as well as from internally deposited radionuclides.

Agreement State: Certain states have entered into an agreement with the NRC to license and regulate the use of radioactive materials within the state. The state regulations in agreement states must be compatible with and at least as stringent as NRC regulations.