ESTIMATION OF DOSES TO INDIVIDUALS FROM RADIONUCLIDES DISPOSED OF IN SOLID WASTE STORAGE AREA 6

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ABSTRACT

A simple methodology has been applied to estimate maximum possible doses to individuals from exposure to radionuclides released from Solid Waste Storage Area No. 6. This is the only operating shallow-land disposal site for radioactive waste at the Oak Ridge National Laboratory. The methodology is based upon simple, conservative assumptions. A data base of radionuclides disposed of in trenches and auger holes was prepared, and several radionuclide transport and ingestion scenarios were considered. The results of these simulations demonstrate the potential for adverse health effects associated with this waste disposal area, and support the need for further calculations using more complete and realistic assumptions.

INTRODUCTION

The radioactive waste disposal area designated Solid Waste Storage Area No. 6 (SWSA6) is the only operating shallow-land burial site at Oak Ridge National Laboratory (ORNL). Burial was initiated on this site on a limited scale in 1969[1], with full operations beginning in 1973. Net yearly disposal inventory values are available for years 1971-76, while radionuclide-specific values are available for 1977-84. By the end of 1984, about 29,000 m^3 of low level waste, containing (at time of burial) 251,000 Ci of radioactivity had been disposed on SWSA6, occupying an area of 3.5 ha. These wastes were classified as either general radioactive waste (this category further subdivided into two subclasses, depending upon radiation level at the surface of the container), mixed wastes, or low-hazard contaminated waste. Other waste categories, not disposed of in SWSA6, were U-233/transuranium waste and U-235 waste.

As a first step in the systematic investigation of the possibility of future exposure of local individuals from radionuclides released from SWSA6, a simple methodology was devised and applied to generate conservative estimates of possible doses resulting from exposures via the hydrologic pathway. For the Oak Ridge area, it is thought that this pathway is the most significant for long-term radionuclide release and transport.

We have completed the initial estimations of the potential doses associated with ingesting ground water and river water that may be contaminated by leachate from SWSA6. The purposes of this activity were to (a) identify which radionuclides might pose future health hazards, and which should be considered in a more thorough pathways analysis; (b) provide preliminary, order-of-magnitude estimates of effective dose equivalent values for a single important exposure pathway; (c) help identify possible deficiencies in the available data for SWSA6; and (d) support investigations as to which environmental parameters might contribute the most uncertainty to dose estimates of human exposure associated with the SWSA6 activities.

TASK DESCRIPTION

Dose estimates were calculated for 35 radionuclides using inventory data for (fiscal) years 1977-1984. All radionuclide inventory data are taken from the Integrated Data Base[2], which is part of the SWMS data base. The SWMS data base has been described elsewhere[3]. These values were "corrected", as described below, to account for radionuclides disposed of during the period 1971-1976, during which detailed, radionuclide-specific inventory data were not recorded. Trench and auger-hole inventories and releases were considered separately in these simulations. These inventory data for years 1977-84 are summarized in Tables I and II. Also considered was ingestion of water taken either from shallow wells or from the Clinch River following dilution by the river flow. Radionuclide-specific dose estimates were calculated following radioactive decay for periods of 100, 1000, and 10^4 years[4].

METHODOLOGY

The radionuclide-specific waste inventory for years 1977-84 is first augmented to account for disposal during years 1971-76. Letting the radionuclide-specific and the total radionuclide activity inventory for years 1971-76 be RA(71-76) and TI(71-76), and with similar definitions for years 1977-84 for which radionuclide-specific records were kept, the assumption that the distribution of radionuclide activities for the earlier period is represented by that for the latter period leads to

$$RA(71\text{-}76) = RA(77\text{-}84) * TI(71\text{-}76) / TI(77\text{-}84).$$

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A more useful expression, with similar definitions for the period 1971-84, is

RA(71-84) = RA(77-84) * <1 + TI(71-76) / TI(77-84)>,

The correction factor within the braces will be referred to as FC. It was assumed that the radionuclide-specific activity that is released in a given year is equal to the activity of

TABLE I

Inventory of radionuclides disposed of in trenches SWSA6 during fiscal years 1977-84.

Nuclide	Amount(Ci)
H-3	4314
Be-10	.2031
C-14	251.5
Na-22	.5411
Fe-55	.755
Co-60	9943
Ni-63	.001
Sr-90	250.8
Zr-93	5.112
Tc-99	35.16
Ru-106	10.05
Cd-113m	.0025
Sn-121m	.5
Cs-134	4.427
Cs-137	644.9
Pm-147	59.7
Sm-151	3.249
Eu-152	5.62
Eu-154	3.34
Eu-155	.05
Np-237	.000449
Th-232	2.564
U-233	227.1
U-235	5.508
U-238	204.18
Pu-238	.027
Pu-239	.133
Pu-240	0
Pu-242	.00499
Am-241	3.438
Am-243	.0000399
Cm-242	.000005
Cm-244	5.583
Bk-249	.000001
Cf-252	.0092136
Unidentified	1625.3

radionuclide originally buried during the years 1971-84, times a (radionuclide-specific) radionuclide decay factor computed from the half-life TH, times a release factor FR. Thus, a radionuclide-specific annual release RR for the year of interest, after an elapsed time T, is

$$RR = RA(77\text{-}84) * e^{(-.693\ T/TH)} * FC * FR.$$

The activity of radionuclide ingested will not in general be as large as that released, since the radionuclide will be diluted into some volume of water that is usually much greater than a human will ingest as water in one year. Thus, the radionuclide-specific activity ingested is computed by multiplying the annual release by a dilution factor FL for water ingestion. The dilution factor is computed from the original volume of waste leachate that is specified by the modeler as being released in the year of interest and from the assumed yearly volume of water ingested by a human (assumed 370 liters). The waste leachate volumes were calculated for the SWSA6 site, assuming that the infiltration percolates vertically through the waste, by multiplying the annual infiltration average for local watersheds (48 cm) by the net horizontally projected area of the trenches or auger-holes to obtain values of 3960 m^3/y for the trenches and 206 m^3/y for the auger-holes. Only the auger-holes and trenches identified as

TABLE II

Inventory of radionuclides disposed of in auger holes on SWSA6 during fiscal years 1977-84.

Nuclide	Amount(Ci)
H-3	2791
Be-10	400
C-14	.000001
Na-22	0
Fe-55	10
Co-60	22140
Ni-63	0
Sr-90	2719
Zr-93	18.51
Tc-99	.612
Ru-106	0
Cd-113m	3
Sn-121m	9
Cs-134	0
Cs-137	3651
Pm-147	85
Sm-151	199.6
Eu-152	50850
Eu-154	72600
Eu-155	31310
Np-237	.0000003
Th-232	.08338
U-233	0
U-235	.014
U-238	.444
Pu-238	.0019
Pu-239	.0000103
Pu-240	0
Pu-242	0
Am-241	.0001499
Am-243	1.059E-05
Cm-242	0
Cm-244	0
Bk-249	0
Cf-252	0
Unidentified	5614.2

containing radionuclide wastes were considered for this calculation. For the scenario of river water ingestion, an annual flow value for the Clinch River of 4.2 (10^9) m^3/y was used.

For this simple calculation of possible individual doses, certain considerations were neglected. No consideration was made of additional leachate dilution en route to a well. No consideration was made of retardation of radionuclides by chemical exchange with soil or other waste material. If this consideration had been made, it would have been appropriate also to examine the consequences of the inclusion of complexing and chelating agents. No consideration was made of daughter ingrowth prior to ingestion of the parent radionuclide. No consideration was made of radionuclide recycling within the watershed.

Dose conversion factors from ICRP-30[4] were used to convert annual radionuclide ingestion values to 50-y committed effective dose equivalent values. No account was taken that an individual might drink well or river water during each of several years and thereby accrue a larger dose commitment.

It should be noted that a release factor is not the same as a release rate. A release factor of value FR indicates that a fraction FR of the decayed radionuclide activity escaped confinement in the year of interest; therefore, it is impossible using this simple model to sustain a specified release fraction FR for greater than 1/FR years. More complete simulations of release, transport, and exposure are now being performed using the PRESTO-II model[5], and these simulations include the determination of realistic time-dependent release rates, inclusion of multiple exposure pathways, and evaluation of exposure individuals of different age.

RESULTS

The numerical results are summarized in Tables III-VI. Tables III and IV summarize the computed maximum exposures to individuals drinking ground water contaminated by leachate from the SWSA6 trenches and wells, while Tables V and VI consider the additional dilution that would result if contaminated ground water flowed into the Clinch River, from which water would be taken and ingested.

Tables III and IV show that several of the dose estimates for groundwater ingestion are high (above .5 rem/y) for all the periods examined. Radionuclides falling into this category include Be-10, Th-232, and isotopes of U. Whereas the Be-10 is disposed of in known auger-holes, the locations of Th- and U-containing wastes are less easily localized. Examination of simulation results to identify the isotopes with potentially problem doses (greater than 25 mrem/y) for the groundwater ingestion pathway after an elapsed time of 100 y suggests a number of radionuclides that should be considered further. Radionuclides of concern that were disposed of in trenches are C-14, Sr-90, Tc-99, Cs-137, Th-232, U-233, U-235, U-238, Pu-239, Am-241, and Cm-244. Radionuclides of concern that were disposed of in auger-holes are Be-10, Co-60 (.024 rem/y), Sr-90, Zr-93, Tc-99 (0.019 rem/y), Cd-113, Sn-121M, Th-232, U-235, and U-238. In summary, radionuclides that are being further examined in a more formal pathways/dose estimation exercise covering periods greater than 100 y are Be-10, C-14, Co-60, Sr-90, Sr-93, Tc-99, Cd-113, Sn-121, Cs-137, Th-232, U-233, U-235, U-238, Pu-239, Am-241, and Cm-244. Still other radionulcides may need to be considered for other pathways.

Contamination of local ground and river water may occur considerably before 100 y has elapsed, and well water from the environs of SWSA6 is being monitored[2] to identify any possible migration of contamination plumes.

Tables V and VI do not demonstrate doses considered significant for individual exposure, but such calculations are of value for estimating population doses in consideration of ALARA.

UNCERTAINTIES

A major uncertainty exists in lack of knowledge of the activities of radionuclides disposed of on SWSA6 during the years 1971-76. The activity disposed of during this period is about 19% of the total activity, an amount that was accounted for in this calculation, but the radionuclide-specific uncertainty associated with this lack of information may be large. Since many factors other than activity are important in evaluating dose commitments, the calculated dose values may contain significant error as a result of the uncertainty in the inventory values. We may be able to estimate the potential inventory uncertainty by examining shipping records or interviewing waste providers or disposal personnel. Additional inventory

TABLE III

Estimated maximum possible doses to an individual ingesting contaminated groundwater containing radionuclides originating in SWSA6 trenches, for various values of elapsed time T. These estimates were calculated using the simple, conservative methodology discussed in the text. For all results discussed here, a release factor of 0.01 was assumed.

Maximum Individual Dose (Rem)

Nuclide Name	Elapsed Time (y) 100	1000	10^4
H-3	.11E-02	.10E-24	.00E+00
Be-10	.11E-02	.11E-02	.11E-02
C-14	.58E+00	.52E+00	.17E+00
Na-22	.18E-13	.00E+00	.00E+00
Fe-55	.14E-14	.00E+00	.00E+00
Co-60	.57E-03	.00E+00	.00E+00
Ni-63	.32E-06	.63E-09	.00E+00
Sr-90	.34E+01	.78E-09	.00E+00
Zr-93	.95E-02	.95E-02	.94E-02
Tc-99	.57E-01	.57E-01	.55E-01
Ru-106	.00E+00	.00E+00	.00E+00
Cd-113	.27E-05	.33E-25	.00E+00
Sn-121	.25E-03	.29E-08	.00E+00
Cs-134	.75E-15	.00E+00	.00E+00
Cs-137	.36E+01	.33E-08	.00E+00
Pm-147	.23E-12	.00E+00	.00E+00
Sm-151	.64E-03	.49E-06	.00E+00
Eu-152	.13E-03	.34E-26	.00E+00
Eu-154	.47E-03	.55E-20	.00E+00
Eu-155	.20E-20	.00E+00	.00E+00
Np-237	.20E-01	.20E-01	.20E-01
Th-232	.78E+01	.78E+01	.78E+01
U-233	.73E+02	.73E+02	.70E+02
U-235	.16E+01	.16E+01	.16E+01
U-238	.58E+02	.58E+02	.58E+02
Pu-238	.53E-02	.39E-05	.00E+00
Pu-239	.65E-01	.64E-01	.49E-01
Pu-240	.00E+00	.00E+00	.00E+00
Pu-242	.23E-02	.23E-02	.23E-02
Am-241	.72E+01	.19E+01	.23E-05
Am-243	.96E-04	.89E-04	.38E-04
Cm-242	.00E+00	.00E+00	.00E+00
Cm-244	.14E+00	.57E-16	.00E+00
Bk-249	.00E+00	.00E+00	.00E+00
Cf-252	.23E-13	.00E+00	.00E+00

uncertainty is derived from the presence of unidentified radionuclides in the 1977-84 records. The error in total inventory is only about 3%; yet, again, the error in specific radionuclide activity values may be large. These uncertainties might be reduced in some cases if a ground-surface survey were made of SWSA6 using a gamma detector and multichannel analyzer. Analyses of ground water and trench leachate might be of value here, but results would be heuristic rather than conclusive.

A complete and defensible pathways analysis should consider retardation of radionuclides by chemical exchange. Radionuclide-specific K_d values

TABLE IV

Estimated maximum possible doses to an individual ingesting contaminated groundwater containing radionuclides originating in SWSA6 auger holes, for various values of elapsed time T. These estimates were calculated using the simple, conservative methodology discussed in the text.

Maximum Individual Dose (Rem)

Nuclide Name	Elapsed Time (y) 100	1000	10^4
H-3	.13E-01	.13E-23	.00E+00
Be-10	.40E+02	.40E+02	.40E+02
C-14	.44E-07	.40E-07	.13E-07
Na-22	.00E+00	.00E+00	.00E+00
Fe-55	.35E-12	.00E+00	.00E+00
Co-60	.24E-01	.00E+00	.00E+00
Ni-63	.00E+00	.00E+00	.00E+00
Sr-90	.71E+03	.16E-06	.00E+00
Zr-93	.66E+00	.66E+00	.66E+00
Tc-99	.19E-01	.19E-01	.19E-01
Ru-106	.00E+00	.00E+00	.00E+00
Cd-113	.63E-01	.77E-21	.00E+00
Sn-121	.85E-01	.10E-05	.00E+00
Cs-134	.00E+00	.00E+00	.00E+00
Cs-137	.39E+03	.36E-06	.00E+00
Pm-147	.62E-11	.00E+00	.00E+00
Sm-151	.75E+00	.58E-03	.00E+00
Eu-152	.22E+02	.59E-21	.00E+00
Eu-154	.20E+03	.23E-14	.00E+00
Eu-155	.24E-13	.00E+00	.00E+00
Np-237	.25E-03	.25E-03	.25E-03
Th-232	.49E+01	.49E+01	.49E+01
U-233	.00E+00	.00E+00	.00E+00
U-235	.80E-01	.80E-01	.80E-01
U-238	.24E+01	.24E+01	.24E+01
Pu-238	.72E-02	.53E-05	.00E+00
Pu-239	.97E-04	.95E-04	.73E-04
Pu-240	.00E+00	.00E+00	.00E+00
Pu-242	.00E+00	.00E+00	.00E+00
Am-241	.61E-02	.16E-02	.19E-08
Am-243	.49E-03	.45E-03	.19E-03
Cm-242	.00E+00	.00E+00	.00E+00
Cm-244	.00E+00	.00E+00	.00E+00
Bk-249	.00E+00	.00E+00	.00E+00
Cf-252	.00E+00	.00E+00	.00E+00

should be measured using actual trench or auger-hole leachate and soils from various depths of SWSA6. K_d values are site- and soil- specific, and literature estimates using average values may be in error by orders of magnitude.

Ground water velocities (speed and direction) need to be measured to permit evaluation of transit time to the site boundary. If ground water velocity is low, radionuclide concentrations in ground water would likely exceed those values calculated in this exercise.

Waste chemistry in the trenches or auger-holes and during transit could have a significant influence on chemical retardation and transport velocity. Leachate and ground water sampling would contribute to our understanding of transport mechanisms as influenced by chemical specification.

Waste chemistry is a function of other waste components, and by site soils and ground water.

TABLE V

Estimated maximum possible doses to an individual ingesting contaminated river water containing radionuclides originating in SWSA6 trenches, for various values of elapsed time T. These estimates were calculated using the simple, conservative methodology discussed in the text.

Maximum Individual Dose (Rem)

Nuclide Name	Elapsed Time (y) 100	1000	10^4
H-3	.10E-08	.97E-31	.00E+00
Be-10	.10E-08	.10E-08	.99E-09
C-14	.55E-06	.49E-06	.16E-06
Na-22	.17E-19	.00E+00	.00E+00
Fe-55	.13E-20	.00E+00	.00E+00
Co-60	.53E-09	.00E+00	.00E+00
Ni-63	.30E-12	.60E-15	.00E+00
Sr-90	.32E-05	.73E-15	.00E+00
Zr-93	.89E-08	.89E-08	.89E-08
Tc-99	.54E-07	.54E-07	.52E-07
Ru-106	.00E+00	.00E+00	.00E+00
Cd-113	.26E-11	.31E-31	.00E+00
Sn-121	.23E-09	.28E-14	.00E+00
Cs-134	.71E-21	.00E+00	.00E+00
Cs-137	.34E-05	.31E-14	.00E+00
Pm-147	.21E-18	.00E+00	.00E+00
Sm-151	.60E-09	.46E-12	.00E+00
Eu-152	.12E-09	.32E-32	.00E+00
Eu-154	.44E-09	.52E-26	.00E+00
Eu-155	.19E-26	.00E+00	.00E+00
Np-237	.19E-07	.19E-07	.19E-07
Th-232	.74E-05	.74E-05	.74E-05
U-233	.69E-04	.69E-04	.66E-04
U-235	.15E-05	.15E-05	.15E-05
U-238	.55E-04	.55E-04	.55E-04
Pu-238	.50E-08	.37E-11	.00E+00
Pu-239	.61E-07	.60E-07	.46E-07
Pu-240	.00E+00	.00E+00	.00E+00
Pu-242	.22E-08	.22E-08	.22E-08
Am-241	.68E-05	.18E-05	.21E-11
Am-243	.91E-10	.84E-10	.36E-10
Cm-242	.00E+00	.00E+00	.00E+00
Cm-244	.13E-06	.54E-22	.00E+00
Bk-249	.00E+00	.00E+00	.00E+00
Cf-252	.21E-19	.00E+00	.00E+00

Depth of waste cavities (trenches or auger-holes), distances from cavity bottoms to the saturated zone as a functions of time, aquifer depth and dispersion coefficient, aquifer terminus, surface water transport rates and flow patterns, and magnitudes of water influx to the waste cavity through the tops and sides are physical parameters describing hydrologic transport that are relevant to narrowing the ranges of uncertainties of waste leaching and transport.

Leachability or diffusion parameters for concrete containers and deterioration and degradation of these and other waste containers and poured containment material would be helpful. In some cases, superficial yet perhaps helpful container descriptions are recorded in disposal records.

TABLE VI

Estimated maximum possible doses to an individual ingesting contaminated river water containing radionuclides originating in SWSA6 auger holes, for various values of elapsed time T. These estimates were calculated using the simple, conservative methodology discussed in the text.

Maximum Individual Dose (Rem)

Nuclide Name	Elapsed Time (y) 100	1000	10^4
H-3	.66E-09	.63E-31	.00E+00
Be-10	.20E-05	.20E-05	.20E-05
C-14	.22E-14	.19E-14	.66E-15
Na-22	.00E+00	.00E+00	.00E+00
Fe-55	.17E-19	.00E+00	.00E+00
Co-60	.12E-08	.00E+00	.00E+00
Ni-63	.00E+00	.00E+00	.00E+00
Sr-90	.35E-04	.79E-14	.00E+00
Zr-93	.32E-07	.32E-07	.32E-07
Tc-99	.94E-09	.94E-09	.91E-09
Ru-106	.00E+00	.00E+00	.00E+00
Cd-113	.31E-08	.38E-28	.00E+00
Sn-121	.42E-08	.50E-13	.00E+00
Cs-134	.00E+00	.00E+00	.00E+00
Cs-137	.19E-04	.18E-13	.00E+00
Pm-147	.30E-18	.00E+00	.00E+00
Sm-151	.37E-07	.28E-10	.00E+00
Eu-152	.11E-05	.29E-28	.00E+00
Eu-154	.96E-05	.11E-21	.00E+00
Eu-155	.12E-20	.00E+00	.00E+00
Np-237	.12E-10	.12E-10	.12E-10
Th-232	.24E-06	.24E-06	.24E-06
U-233	.00E+00	.00E+00	.00E+00
U-235	.39E-08	.39E-08	.39E-08
U-238	.12E-06	.12E-06	.12E-06
Pu-238	.35E-09	.26E-12	.00E+00
Pu-239	.48E-11	.46E-11	.36E-11
Pu-240	.00E+00	.00E+00	.00E+00
Pu-242	.00E+00	.00E+00	.00E+00
Am-241	.30E-09	.76E-10	.93E-16
Am-243	.24E-10	.22E-10	.95E-11
Cm-242	.00E+00	.00E+00	.00E+00
Cm-244	.00E+00	.00E+00	.00E+00
Bk-249	.00E+00	.00E+00	.00E+00
Cf-252	.00E+00	.00E+00	.00E+00

Laboratory measurements that might provide valuable quantification of processes related to radionuclide transport include leaching tests, chemical exchange parameter measurements using typical waste components to accurately replicate the chemical environment of the trench or auger hole, diffusion and degradation tests of container and poured containment materials, and leachate analysis.

WORK IN PROGRESS

A data set containing environmental and radiological parameters for SWSA6 has been prepared, and simulations are being performed using the PRESTO-II model[5]. These simulations are a more realistic pathways analysis and radiation dose calculation for wastes disposed of on SWSA6. A more complete set of exposure scenarios is being considered, and calculations are being made of exposures to site intruders, to residents at the site boundary, and to individuals using well and river water for a variety of applications. Input parameters will be varied over a reasonable range of values to examine the effects of uncertainties in input parameters on predicted radiation exposures.

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