Review of Radionuclide Source Terms used for Performance-Assessment Analyses

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Abstract

Two aspects of the radionuclide source terms used for total-system performance-assessment (TSPA) analyses have been reviewed. First, a detailed radionuclide inventory (i.e., one in which the reactor type, decay, and burnup are specified) is compared with the standard source-term inventory used in prior analyses. The latter assumes a fixed ratio of pressurized-water reactor (PWR) to boiling-water reactor (BWR) spent fuel, at specific amounts of burnup and at 10-year decay. TSPA analyses have been used to compare the simplified source term with the detailed one. The TSPA-91 analyses did not show a significant difference between the source terms.

Second, the radionuclides used in source terms for TSPA aqueous-transport analyses have been reviewed to select ones that are representative of the entire inventory. It is recommended that two actinide decay chains be included (the $4n+2$ "uranium" and $4n+3$ "actinium" decay series), since these include several radionuclides that have potentially important release and dose characteristics. In addition, several fission products are recommended for the same reason. The choice of radionuclides should be influenced by other parameter assumptions, such as the solubility and retardation of the radionuclides.

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"The data in this report were developed subject to QA
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1.0 Introduction

The recently completed preliminary total-system performance assessment for the potential Yucca Mountain waste repository (TSPA-91) (Barnard *et al.*, 1992) included four components, representing various release mechanisms. The processes modeled included aqueous transport (due to "nominal" geohydrologic conditions and due to human-intrusion activities), gaseous transport, and direct surface release due to human intrusion and igneous activity. The analyses and assumptions for TSPA-91 are discussed in detail in Barnard *et al.* (1992) and will not be repeated here. The source term for direct surface releases differed somewhat from those for aqueous releases because of different assumptions about transport. The aqueous analyses assumed that not all radionuclides in the inventory would reach the accessible environment because of differing solubility and retardation properties of the radionuclides. For direct surface releases, the transport mechanisms were assumed to be mechanical, so the same differentiation would not occur.

This report discusses two aspects of TSPA source terms; the first is studies done for the TSPA-91 to evaluate the sensitivity of some of the calculated direct-surface releases to the choice of source-term inventory. Second, it reviews the choice of radionuclides used in the TSPA-91 source terms and evaluates which ones should be used in future analyses to be most representative of the repository inventory.

2.0 Radionuclide Inventories

The radionuclide source terms used in these analyses were based on the design radionuclide inventory, as specified in the Yucca Mountain Site Characterization Plan (SCP) (DOE, 1988a, Chapter 7). A source term based on this inventory would have characteristic thermal and radioactive decay profiles, as well as characteristic mobilization behavior, resulting from the combination of radionuclides included. Changing any of the parameters of the source term may change its decay and mobilization characteristics. Such changes could affect the amount and timing of radionuclide releases.

The radionuclide inventory present in spent fuel is a function of several factors—burnup, decay, and reactor type. Burnup of nuclear fuel is expressed as the fission energy released as a function of the initial amount of uranium in the fuel (given in megawatt-days per metric ton of fissile uranium—MWd/MTU). The buildup of fission products and reaction-product actinides in nuclear fuel is proportional to the number of fissions that have occurred (i.e., the burnup). After fuel is discharged from the reactor, essentially no new fission products are created, and the

radioactive nuclides begin to decay. As the radionuclides decay, there is an increase in the inventory of some isotopes that are daughter products of other isotopes. The fuels used in pressurized-water reactors (PWRs) and that used in boiling-water reactors (BWRs) are substantially the same in amount of uranium per fuel assembly. Because PWR fuel is designed to have higher ^{235}U enrichment, it is generally irradiated to a greater burnup than BWR fuel.

The standard inventory (as discussed below) represents simplifications and averages of the actual inventory of radionuclides. Developing descriptions of the spent-fuel inventory that more accurately describe the decay and burnup of the entire inventory is not difficult. This report does so, and evaluates whether the extra effort involved in including this information in performance-assessment analyses would make a difference in our interpretation of those analyses.

Although the baseline design of the potential repository calls for an inventory of both light-water reactor (LWR) spent fuel and defense high-level waste (i.e., glassified waste), only the former type of waste is considered here.

2.1 Radionuclides Included in Source Terms

Although spent nuclear fuel contains many radionuclides, some have such short half-lives, and others have such low inventories that they probably do not represent contamination threats when they are emplaced in a repository. The EPA has specified 20 years as the lower cutoff for radionuclide half-lives that are considered to be of regulatory concern. They have published release limits in 40 CFR Part 191, Appendix A (EPA, 1985) that specify the maximum fractional amounts (per MTU of repository waste) of radionuclides that may be released over 10,000 years. (40 CFR 191 has been remanded, but until new regulations are issued, these regulations will continue to be one of the measures of performance-assessment analyses).

Using these criteria of half-life, inventory and EPA release limit, a list of radionuclides potentially contributing to releases has been developed (Wilson, 1991). Table 1 gives the list of radionuclides, their half-lives, and the EPA limits. Actinides are listed first, grouped by decay chain, followed by the fission products (shaded in the table). Radionuclides from this table were used in the TSPA-91 analyses, as indicated by the footnotes to the table.

Table 1
Radioisotopes used in Source Terms for TSPA-91

Isotope	Half-life (years)	EPA Limit (Ci/MTU)	Inventory (Ci/MTU)
^{246}Cm	4.731×10^3	0.1	2.58×10^{-2}
^{242}Pu	3.869×10^5	0.1	1.60×10^0
^{238}U	4.468×10^9	0.1	3.18×10^{-1}
^{242}Am	1.520×10^2	0.1	7.46×10^0
^{238}Pu	8.774×10^1	0.1	2.12×10^3
$^{234}\text{U}^{\dagger\dagger}$	2.445×10^5	0.1	1.13×10^0
^{230}Th	7.700×10^4	0.01	1.29×10^{-4}
^{226}Ra	1.600×10^3	0.1	3.67×10^{-7}
^{210}Pb	2.230×10^1	1.0	4.71×10^{-8}
^{243}Cm	2.850×10^1	0.1	1.54×10^1
$^{243}\text{Am}^{\dagger\dagger}$	7.380×10^3	0.1	1.55×10^1
$^{239}\text{Pu}^{\dagger\dagger}$	2.406×10^4	0.1	3.08×10^2
^{235}U	7.038×10^8	0.1	1.68×10^{-2}
^{231}Pa	3.277×10^4	0.1	1.94×10^{-5}
^{227}Ac	2.177×10^1	0.1	5.19×10^{-6}
^{245}Cm	8.499×10^3	0.1	1.26×10^{-1}
^{241}Pu	1.440×10^1	0.0	7.43×10^4
$^{241}\text{Am}^{\dagger}$	4.322×10^2	0.1	1.64×10^3
$^{237}\text{Np}^{\dagger\dagger}$	2.140×10^6	0.1	2.88×10^{-1}
^{233}U	1.585×10^5	0.1	2.54×10^{-5}
^{229}Th	7.339×10^3	0.1	1.40×10^{-7}
^{244}Cm	1.811×10^1	0.0	1.15×10^3
$^{240}\text{Pu}^{\dagger}$	6.537×10^3	0.1	5.38×10^2
^{236}U	2.341×10^7	0.1	2.40×10^{-1}
^{232}U	7.200×10^1	0.1	2.50×10^{-2}
^{151}Sm	8.999×10^1	1.0	3.18×10^2
^{137}Cs	3.000×10^1	1.0	7.66×10^4
$^{135}\text{Cs}^{\dagger\dagger}$	2.300×10^6	1.0	3.50×10^{-1}
$^{129}\text{I}^{\dagger\dagger}$	1.570×10^7	0.1	2.95×10^{-2}

Table 1, continued
Radioisotopes used in Source Terms for TSPA-91

Isotope	Half-life (years)	EPA Limit (Ci/MTU)	Inventory (Ci/MTU)
$^{126}\text{Sn}^{\dagger\dagger}$	1.000×10^5	1.0	7.17×10^{-1}
^{121}Sn	4.997×10^1	1.0	9.04×10^{-1}
^{108}Ag	1.270×10^2	1.0	1.19×10^{-2}
^{107}Pd	6.496×10^6	1.0	1.05×10^{-1}
$^{99}\text{Tc}^{\dagger\dagger}$	2.130×10^5	10.0	1.23×10^1
^{93}Mo	3.498×10^3	1.0	1.60×10^{-2}
^{94}Nb	2.030×10^4	1.0	7.93×10^{-1}
^{93}Zr	1.530×10^6	1.0	1.88×10^0
^{90}Sr	2.912×10^1	1.0	5.32×10^4
$^{79}\text{Se}^{\dagger\dagger}$	6.496×10^4	1.0	3.81×10^{-1}
^{63}Ni	9.200×10^1	1.0	4.55×10^2
^{59}Ni	8.000×10^4	1.0	3.56×10^0
^{36}Cl	3.010×10^5	1.0	1.19×10^{-2}
$^{14}\text{C}^{\dagger*}$	5.729×10^3	0.1	1.54×10^0

† Isotopes used for the saturated-zone source term.

†† Isotopes used for the aqueous-transport source term.

* Isotope used for the gaseous-transport source term.

The radionuclides used for the source terms for TSPA-91 were chosen to be representative of the entire nuclear-waste inventory. Different numbers of radionuclides were included in the source terms for the three types of releases (aqueous, gaseous, and surface-direct), depending on the nature of the assumed release processes. For aqueous releases, only radionuclides which were thought to be readily mobilized and transported in groundwater were included. For gas-flow analyses, only CO_2 transport was considered, so only ^{14}C was used in that source. Because direct release was assumed to occur by mechanical means, all radionuclides listed in Table 1 were included. The radionuclides used in each type of analysis are indicated in the table.

3.0 Description of Source Terms for Direct-Surface Releases

Source term inventories for TSPA-91 have been constructed from the Nuclear Waste Terminal Storage Program (NWTSP) database (Roddy *et al.*, 1986). This database provides activities for radionuclides as a function of decay, burnup, and reactor type. It was constructed from predictions of radionuclide inventories from

reactor operations produced by the ORIGEN2 code (Croff, 1980). The ORIGEN2 simulations were based on typical values for LWR fuel-assembly physical characteristics, structural-material composition, and nonactinide and actinide compositions of the oxide fuel. The values are listed in Tables 3.1 - 3.4 of Roddy *et al.* (1986).

The standard source term uses ORIGEN data for burnup values specified in the SCP at 10 years' decay, weighted by the assumed proportion of PWR fuel to BWR fuel. The detailed source terms have been constructed using additional data from the *LWR Characteristics Database* (DOE, 1992, v. 1), providing a wider range of burnup and decay values. This work is an update and enhancement of the NWTSP database.

3.1 Standard Source Term

The waste acceptance criteria in the SCP are based on the definition given in 10 CFR Part 961 for Spent Nuclear Fuel and High-Level Radioactive Wastes (DOE, 1987). This specifies that spent LWR fuel will have decayed a minimum of 5 years since discharge from the reactor. The SCP assumed that the potential Yucca Mountain repository would receive waste starting in 1998 and continue through 2022. Using those assumptions, the LWR spent fuel would initially be considerably older than 10 years, but would approach the 5-year limit toward the end of the loading cycle of the repository.

The ^{235}U enrichment for PWRs is about 3.2 weight percent, compared to about 2.6 percent for BWRs. At these enrichments, average burnups are 33,000 MWd/MTU for PWR fuel and 27,500 MWd/MTU for BWR fuel. As reactor technologies have matured, the average burnup of the fuel has increased. Thus, one should expect the average burnup of fuel received later in the repository loading phase to be greater than the burnup of the older fuel.

The last factor in the standard source term is the proportion of PWR fuel to BWR fuel. The SCP (Tables 6-8 and 7-2) indicates that roughly 54% of the total nuclear waste in the repository will be spent PWR fuel, 35% will be BWR spent fuel, and the balance will be defense high-level waste. To simplify the analyses for the TSPA-91, the high-level defense waste has been ignored, resulting in a standard source term consisting of 60% PWR fuel with a burnup of 33,000 MWd/MTU, and 40% BWR fuel with a burnup of 27,500 MWd/MTU. In Table 1, the "Inventory" column lists the activities for the standard source term. Note that ^{241}Pu and ^{244}Cm are included in the source term, even though EPA limits have not been specified (because of their short half-lives). They have large inventories, and they are parent members of important radionuclides below them in the respective decay chains.

3.2 Detailed Source Term

The detailed source term incorporates two changes in assumptions from the standard source. First, since the time of operation of the potential repository has slipped from a first receipt of waste in 1998 by about 20 years, there will be a much larger amount of spent fuel that has decayed for much longer than 10 years. Second, for some of the nonaqueous-flow analyses in TSPA-91, the results can depend directly on the nature of the radionuclide inventory. The detailed source term was initially developed for the human-intrusion component of the TSPA-91 analysis. For this analysis, releases occur directly at the surface above the repository because of drilling operations. Under these circumstances, the exact inventory of the radionuclides can have a direct bearing on the releases. Because the human-intrusion analysis is treating a non-normal condition at the repository, the decay time for the radionuclides is based on closure of the repository in the year 2040. (It is exceedingly unlikely that there would be inadvertent drilling into the repository while activities are occurring there.)

Using inventory information from the LWR Characteristics Database, the amount of spent fuel that will be discharged from light-water nuclear reactors from the beginning of commercial power production (1968 for BWRs and 1970 for PWRs) through the forecast end of their use (approximately 2040) can be estimated. As the technology of nuclear power has changed, the amount of burnup of the fuel has increased, from an average of about 10,000 MWd/MTU for 1970s-vintage BWRs to a projected average of over 45,000 MWd/MTU for future PWRs. Figure 1 shows the distribution of reactor spent-fuel inventories as a function of burnup and year of discharge for PWRs. (Discharges from BWRs have a similar distribution.) Tables 2 and 3 give the actual and projected amounts of spent fuel discharged from PWRs and BWRs, respectively. (These tables are adapted from the LWR Characteristics Database, and they update the data in Table 7-2 of the SCP.) These amounts are used to calculate radionuclide inventories weighted by burnup and decay.

Figure 1 and Table 2 show that the peak discharges from PWRs occur in 1990 and 1992, with average burnups of 40,000 MWd/MTU. In 2013 there is predicted to be another large discharge with average burnups greater than 45,000 MWd/MTU. For BWRs there are large discharges of fuel in 1989-1992 and around 2011 (Table 3). Average burnups for the later discharges are also higher than the earlier ones. These data are based on the current DOE scenarios for spent-fuel discharge projections assuming the "No New Orders with Extended Burnup Case" (DOE, 1991). Under these assumptions, existing nuclear reactors (and those under construction) will con-

tinue to operate until their licenses expire (after 40 years). There will be no extensions of the operating licenses. However, the design fuel burnup in those reactors will be gradually increased for PWRs to 55,000 MWd/MTU in 2005 (43,000 MWd/MTU for BWRs). The reactors will continue to operate at those levels until their decommissioning. Nuclear generation capacity is projected to be about 100 gigawatts from 1990 through 2010, and to then decrease to zero by 2030.

Figure 1

Amounts of Spent Fuel Discharged from PWRs

Table 2
Spent-Fuel Inventory Distribution for PWRs (values in MTU)*

— Burnup Bins (MWd/MTU) —

Discharge Year	5000	10000	15000	20000	25000	30000	35000	40000	45000	50000	55000	60000	Total Inventory (MTU)
1970			1.7	37.3									39.0
1971		4.6			2.5	19.4							26.5
1972			11.9	29.3	31.5	23.2	22.1						118.0
1973				26.2		33.3	7.6						67.1
1974		1.5	86.4	13.6	40.5	57.2	1.1						207.7
Totals	7.4	6.1	100.0	106.4	74.5	133.1	30.8						458.3
1975	2.7	42.6	95.0	48.2	84.8	25.3	23.1						321.7
1976			4.6	190.8	82.4	63.3	55.4						396.5
1977			2.8	108.3	118.2	135.2	87.1	15.4					467.0
1978		1.9	48.0	90.3	37.2	339.7	121.9	61.2	0.4				700.6
1979			31.0	107.1	64.0	232.3	234.7	50.1	0.5				719.7
1980			0.4		72.7	238.3	282.4	26.3	2.0		0.9		623.0
1981			17.2	1.9	25.8	230.4	353.0	53.3	1.3				682.9
1982			1.8	81.1	81.4	57.8	297.5	117.4	2.7	0.4	0.4	0.9	641.4
1983		5.5	4.1	71.4	51.1	176.2	321.6	136.2	5.4		0.5		772.0
1984			53.7	45.2	53.1	205.9	378.8	103.9	4.1				844.7
Totals	2.7	50.0	258.6	744.3	670.7	1704.4	2155.5	563.8	16.4	0.4	1.8	0.9	6169.5
1985				44.3	11.8	222.1	330.4	225.1	24.2	0.4			858.3
1986		0.4	27.1	132.5	14.2	190.6	341.3	273.8	35.0	1.3			1017.5
1987			27.6	133.4	82.3	134.2	418.7	334.2	24.4				1154.8
1988				109.6	28.3	139.7	401.8	392.9	84.8	2.1	0.4	2.0	1162.1
1989	0.5	25.9	50.6	126.4	24.9	153.5	337.6	428.5	91.1	11.8		0.4	1250.7
1990			51.3	45.0	10.3	167.8	491.7	599.5	196.3	6.0	0.4		1568.3
1991			25.8		45.5	90.4	268.5	421.0	191.2	24.4			1066.8
1992			30.0	0.4	48.5	91.1	288.6	680.8	302.4	131.1			1572.9
1993			0.4	27.4	26.0	61.5	173.0	346.1	379.9	73.1	3.7		1091.1
1994					2.8	68.0	211.8	488.2	460.2	204.5	3.8		1439.3
Totals	0.5	26.3	212.8	619.0	294.6	1318.9	3263.4	4190.1	1789.5	454.7	9.6	2.4	12181.8

Table 2, Continued

— Burnup Bins (MWd/MTU) —

Discharge Year	5000	10000	15000	20000	25000	30000	35000	40000	45000	50000	55000	60000	Total Inventory (MTU)
1995			0.4	0.5	1.2	84.5	207.3	327.5	473.4	192.7	10.7		1298.2
1996					1.2	0.9	114.4	343.9	415.0	219.7	71.3	0.4	1166.8
1997			0.4			31.6	75.2	380.7	356.5	334.3	108.0	10.4	1297.1
1998			0.4		0.4	27.7	35.5	259.9	361.8	365.9	83.1	54.8	1189.5
1999							25.3	175.6	304.8	428.0	242.4	52.5	1228.6
2000			0.4	8.3			34.7	162.4	300.0	371.3	241.3	73.9	1192.3
2001							13.9	147.6	318.3	422.3	178.8	76.4	1157.3
2002			0.4		25.8		29.9	122.0	245.2	382.3	343.8	56.3	1205.7
2003			0.4			0.3	18.7	195.9	336.0	378.9	159.8	83.1	1173.1
2004							46.1	141.6	242.3	504.0	211.5	42.8	1188.3
Totals			2.4	8.8	28.6	145.0	601.0	2257.1	3353.3	3599.4	1650.7	450.6	12096.9
2005			0.4			6.0	35.9	119.5	417.9	406.7	262.2	65.4	1314.0
2006			0.4				12.3	51.7	350.4	300.3	292.1	118.3	1125.5
2007			19.2	19.0		19.2	0.3	127.2	317.3	313.6	385.5	115.5	1316.8
2008			0.4				7.8	32.9	293.8	303.0	313.1	161.3	1112.3
2009			9.1	2.5		8.8	28.1	32.5	264.3	293.8	335.6	185.4	1160.1
2010			11.6	21.3			33.8	66.8	262.7	300.8	414.9	145.2	1260.8
2011		3.7	38.8	16.3	24.8	49.3	37.4	61.9	400.4	256.1	293.4	102.7	1281.1
2012		5.5		2.9	58.9	11.0	15.5	50.8	312.3	302.3	302.9	137.4	1199.5
2013		8.5	52.9	97.6	39.9	61.4	59.2	154.5	400.4	290.2	347.5	167.3	1679.4
Totals		17.7	132.8	159.6	123.6	155.7	230.3	697.8	3019.5	2766.8	2947.2	1198.5	11449.5
2014			9.7	77.4	36.8	63.7	64.8	99.9	168.7	296.2	294.5	165.6	1277.3
2015			22.3	37.5	20.1	22.3	19.5	82.5	290.0	225.5	130.2	152.5	1002.4
2016	7.6			6.2	99.6	20.0	56.1	51.3	311.3	348.6	223.8	179.2	1303.7
2017				13.3	43.4	67.2	6.7	101.2	143.5	218.9	186.8	116.0	897.0
Totals		7.6	32.0	134.4	199.9	173.2	147.1	334.9	913.5	1089.2	835.3	613.3	4480.4
2018				17.1	11.3		37.5	49.9	217.3	186.7	165.8	95.3	780.9
2019							7.9	45.5	205.3	172.2	137.7	112.5	681.1
2020				14.7	79.1	41.2	45.2	47.2	285.8	194.5	167.3	78.3	953.3
2021				17.3	35.1	16.6	26.3	13.2	212.1	175.1	124.3	71.5	691.5
Totals				49.1	125.5	57.8	116.9	155.8	920.5	728.5	595.1	357.6	3106.8

Table 2, Continued

— Burnup Bins (MWd/MTU) —

Discharge Year	5000	10000	15000	20000	25000	30000	35000	40000	45000	50000	55000	60000	Total Inventory (MTU)
2022			18.6	9.7	21.8	87.0	53.6	60.9	207.6	225.7	138.8	91.2	896.3
2023			18.6	20.4	14.8		37.0	55.1	203.5	67.3	117.5	35.7	569.9
Totals			18.6	30.1	36.6	87.0	90.6	116.0	411.1	293.0	256.3	126.9	1466.2
2024				80.6	41.5	67.4	76.3	34.8	181.7	79.4	147.9	78.4	788.0
2025			27.8	27.9	20.3	63.1	25.9	81.7	148.0	71.1	113.9	45.6	625.3
2026			31.4	32.9	38.8	17.3	73.4	110.0	110.3	68.4	39.0	32.4	553.9
Totals			59.2	141.4	100.6	147.8	175.6	226.5	440.0	218.9	300.8	156.4	1967.2
2027			27.9	52.5	122.1	27.5	72.1	43.2	186.7	104.2	22.0	47.2	705.4
2028			28.1	10.6	53.1	10.6	19.3	28.1	66.1	77.4	12.5	9.3	315.1
2029			28.1		11.6	53.6	18.4	1.0	106.7	15.1	50.4	22.4	307.3
2030			54.9		0.5	29.1	18.8	26.3	62.0	34.4	12.0	9.0	247.0
2031							37.5	48.3	18.6	19.2	5.0	18.5	147.1
2032			25.8		36.9			25.8	73.8	8.3	21.5		192.1
Totals			164.8	63.1	224.2	120.8	166.1	172.7	513.9	258.6	123.4	106.4	1914.0
2033						39.2				10.8	9.8	10.2	70.0
2034								3.0	17.5	6.8			27.3
2035				16.6	20.3		52.2	21.7	15.2	5.3	5.0	18.9	155.2
2036							17.3				4.8	8.9	31.0
Totals				16.6	20.3	39.2	69.5	24.7	32.7	22.9	19.6	38.0	283.5
2037							17.3				4.8	8.9	31.0
2038							35.2				9.7	18.0	62.9
2039							13.6			3.8		6.9	24.3
2040					76.6		18.4	76.6		5.1	21.9	21.3	219.9
Totals					76.6		84.5	76.6		8.9	36.4	55.1	338.1
Grand Totals:	10.6	107.7	981.2	2072.8	1975.7	4082.9	7131.3	8816.0	11410.4	9441.3	6776.2	3106.1	55912.2

* Burnup data adapted from *LWR Characteristics Database* (DOE, 1992, v. 1, Table 2.3).

Table 3
Spent-Fuel Inventory Distribution for BWRs (values in MTU)*

— Burnup Bins (MWd/MTU) —

Discharge Year	5000	10000	15000	20000	25000	30000	35000	40000	45000	50000	55000	60000	Total Inventory (MTU)
1968	0.6												0.6
1969		1.2	1.0	7.3	0.2								9.7
1970	5.6												5.6
1971	41.5	8.1	2.8	10.0	1.6								64.0
1972	97.9	12.1	27.6	4.0									141.6
1973	9.5	16.7	31.0	36.4	1.5	0.1							95.2
1974		78.4	117.7	44.7	3.8								244.6
Totals	155.1	116.5	180.1	102.4	7.1	0.1							561.3
1975	0.3	1.7	62.0	125.6	25.3								214.9
1976	0.9	62.0	105.6	127.7	2.3								298.5
1977		48.0	40.3	235.0	58.9	0.7							382.9
1978	6.3	32.4	13.1	84.2	232.0	15.2							383.2
1979			18.6	108.7	149.2	123.1	0.3						399.9
1980	14.0	0.4	0.6	93.3	413.3	87.6	10.7						619.9
1981		0.2	0.2	58.1	265.4	133.3	0.7	0.7					458.6
1982		0.2	4.6	19.4	144.7	173.6	13.8	0.6	0.4				357.3
1983			0.9	3.0	113.5	337.8	35.7	0.4					491.3
1984		5.4	9.5	0.4	136.2	239.5	70.8		0.4				462.2
Totals	21.5	150.3	255.4	855.4	1540.8	1110.8	132.0	1.7	0.8				4068.7
1985	16.9	42.5	18.3	35.8	77.3	283.9	10.2		0.2				485.1
1986	50.8	38.2	42.6	72.8	62.6	159.4	37.3	0.4					464.1
1987	136.1	36.4	73.2	40.8	108.8	303.1	8.0		0.4				706.8
1988	17.0	24.6	2.4	43.9	176.1	237.4	35.0						536.4
1989	47.8	20.6	97.1	89.0	203.6	214.3	74.4	0.7					747.5
1990	15.7		65.3	50.3	48.3	274.3	175.3	1.3					630.5
1991	0.7			12.5	119.8	218.8	161.5	21.1					534.4
1992			7.1	43.6	106.8	150.5	408.8	21.0					737.8
1993				2.0	8.8	168.5	331.1	45.0					555.4
1994				34.3	10.2	196.3	252.2	131.6					624.6
Totals	285.0	162.3	306.0	425.0	922.3	2206.5	1493.8	221.1	0.6				6022.6

Table 3, Continued

— Burnup Bins (MWd/MTU) —

Discharge Year	5000	10000	15000	20000	25000	30000	35000	40000	45000	50000	55000	60000	Total Inventory (MTU)
1995					14.7	81.9	386.6	185.0	1.4				669.6
1996						76.6	264.5	241.4	34.3				616.8
1997						6.8	136.2	292.5	187.5				623.0
1998						62.0	94.5	237.1	121.2	14.4			529.2
1999						6.9	62.2	369.3	129.2	11.5			579.1
2000						18.2	63.8	349.9	137.4				569.3
2001						2.3	133.5	321.5	176.1	14.2			647.6
2002						16.8	16.2	254.4	222.1	11.7			521.2
2003		2.6		2.6	2.6	0.5	55.4	323.3	238.2	18.1			643.3
2004						10.6	46.6	212.4	232.9	38.0			540.5
Totals	2.6	2.6	2.6	2.6	17.3	282.6	1259.5	2786.8	1480.3	107.9			5939.6
2005							42.9	176.1	278.1	45.5			542.6
2006						10.7	29.4	266.7	244.9	32.8			584.5
2007							30.5	193.7	250.4	39.9			514.5
2008						11.0	41.0	298.0	288.1	27.6			665.7
2009	26.1	56.9	15.4	29.6		57.9	75.9	280.0	238.7	29.9			810.4
2010		33.3	1.4			37.7	44.5	259.1	264.0	27.0			667.0
2011	16.1	63.6	15.4	31.7		47.3	80.3	297.5	289.2	32.0	31.9		905.0
2012		66.2	23.5	31.9		57.8	31.9	125.9	180.0	75.9			593.1
2013		25.9	55.9	14.5		51.0	38.2	138.1	331.4	93.7			748.7
Totals	42.2	245.9	111.6	107.7	107.7	273.4	414.6	2035.1	2364.8	404.3	31.9		6031.5
2014		44.6	182.4	16.8		28.6	196.3	56.0	236.0	140.0	14.6		915.3
2015								56.2	141.1	166.4			363.7
2016		11.6	64.9	2.9		38.5	49.7	17.2	125.0	135.8			445.6
2017								37.1	167.0	106.9			311.0
Totals		56.2	247.3	19.7	19.7	67.1	246.0	166.5	669.1	549.1	14.6		2035.6

Table 3, Continued

— Burnup Bins (MWd/MTU) —

Discharge Year	5000	10000	15000	20000	25000	30000	35000	40000	45000	50000	55000	60000	Total Inventory (MTU)
2018				36.4			36.4	31.5	98.9	145.8			349.0
2019								30.3	80.1	147.3			257.7
2020								36.5	124.0	123.5			284.0
2021								16.6	12.7	59.9	24.4		113.6
Totals				36.4			36.4	114.9	315.7	476.5	24.4		1004.3
2022			20.0	63.7	67.3		63.5	56.6	84.6	283.1			638.8
2023			29.6	46.4	27.5	25.4	46.2	3.5	72.1	78.4			329.1
Totals			49.6	110.1	94.8	25.4	109.7	60.1	156.7	361.5			967.9
2024				40.1			46.9	47.3	155.0	104.8			394.1
2025			9.2	97.5	37.6	12.8	57.2	46.3	105.8	45.5	24.8		436.7
2026			66.5	16.3	45.0	37.9	52.4	55.2	1.4				274.7
Totals			75.7	153.9	82.6	50.7	156.5	148.8	262.2	150.3	24.8		1105.5
2027			31.8	44.8	31.8		48.4	31.8	64.8	31.4			284.8
2029				58.8		38.3		56.6		25.4			179.1
Totals			31.8	103.6	31.8	38.3	48.4	88.4	64.8	56.8			463.9
Grand Totals:	461.6	473.9	1200.7	2148.3	2824.1	4054.9	3896.9	5623.4	5315.0	2106.4	95.7	0.0	28200.9

* Burnup data adapted from LWR Characteristics Database (DOE, 1992, v. 1, Table 2.3).

From an inspection of the tables or the figure, it appears unlikely that a 10-year decay time (as is used in the standard source term) would accurately describe the radionuclide inventory produced over the entire period of reactor operation. Similarly, the burnup values used in the standard source term do not appear to be typical of the entire range.

The NWTSP database lists activities according to reactor type at specific burnup values (5000, 10000, ... MWd/MTU) at specific decay times (1, 2, 3, ..., 15, 20, 25, 30, 40, ..., 100, 200, ... years). The data in Tables 2 and 3 have been grouped by years to match the NWTSP database entries. The weighted average burnups for the groups of decay years have also been calculated, and these values have been rounded to the nearest burnup values found in the database. Thus, instead of a single source term, there are now 22 components that can be used to generate a source term. Table 4 summarizes the values obtained from the LWR Characteristics database. Using the information in Tables 2 and 3, source terms have been built for the specified combinations of reactor type, burnup, and decay. Then depending on the analysis, these source terms have been used in the TSPA-91 analyses.

The shaded entries in Table 4 give the weighted-average burnups for two particular time periods—for the first 25 years of reactor operation (decay years 72 - 46), and for the entire period of operation. The first time period, from 1968-70 through 1994, corresponds to the operational period implied in the SCP analyses. For this period, the weighted-average burnups are fairly close to the standard burnups, but the decay time is much longer than the standard. Over the entire period of operation, the weighted-average burnup values are about 20 percent higher than the ones used in the standard source term.

3.3 Results of Direct-Release Sensitivity Studies

The human-intrusion analysis component of the TSPA-91 used both the standard and detailed source terms. The standard source term assumed that the entire 70,000 MTU of nuclear waste was composed of a 60/40 mixture of PWR and BWR spent fuel. The detailed source term assumed that the oldest waste would be loaded first, and that newer waste would be loaded into the repository until its 70,000-MTU capacity was reached. Table 5 lists the characteristics of the source terms used in TSPA-91. (Note that the burnup values shown in Table 5 differ somewhat from the values in Table 4 as a result of rounding the burnup values for each group differently in the calculation of weighted-average burnups in Tables 2 and 3.)

Table 4
Burnup and Decay Values used for Detailed Source Terms

Reactor Type	Group Range of Decay Years	Decay Years used for Source Term	Group Weighted Average Burnup	Burnup used for Source Term
PWR	70 - 66	70	20683	20000
PWR	65 - 56	60	27665	30000
PWR	55 - 46	50	34740	33000
PWR	45 - 36	40	44298	45000
PWR	35 - 27	30	46342	45000
PWR	26 - 23	25	44709	45000
PWR	22 - 19	20	45414	45000
PWR	18 - 17	18	43214	45000
PWR	16 - 14	15	39715	40000
PWR	13 - 8	10	36784	35000
PWR	7 - 4	5	37931	40000
PWR	3 - 0	2	38167	40000
PWR	70 - 46	—	32077	—
PWR	70 - 0	—	40184	—
BWR	72 - 66	70	10159	10000
BWR	65 - 56	60	22000	20000
BWR	55 - 46	50	25540	25000
BWR	45 - 36	40	37428	35000
BWR	35 - 27	30	37459	35000
BWR	26 - 23	25	37602	40000
BWR	22 - 19	20	43302	45000
BWR	18 - 17	18	35666	35000
BWR	16 - 14	15	33347	35000
BWR	13 - 11	12	31320	30000
BWR	72 - 46	—	23377	—
BWR	72 - 11	—	32028	—

Table 5
Detailed-Source Inventory Parameters used in TSPA-91

Decay Years	% BWR Inventory	BWR Burnup	% PWR Inventory	PWR Burnup	% of Total Inventory
70	55.1	15,000	44.9	25,000	1.46
60	39.7	25,000	60.3	30,000	14.63
50	33.1	27,500	66.9	35,000	26.01
40	32.9	40,000	67.1	45,000	25.77
30	34.5	40,000	65.5	50,000	24.97
25	31.2	40,000	68.8	45,000	7.17

Chapter 6 of the TSPA-91 document discusses in detail the calculations made using the detailed source term. The results are reproduced here in Figure 2. There is essentially no difference between the base case (standard source term) and the analyses done with the detailed source term. Table 5 shows that about 84% of the detailed-source inventory consists of spent fuel with burnups at or above the standard values. The total activity, at time of emplacement, associated with these burnups is given in Table 6 (a 10-year decay is used for the standard burnups, and 40-year decays assumed for all detailed-source burnups).

Figure 2

Comparison of TSPA Results for Standard and Detailed Source Terms

Table 6
Total Radioactivity of Spent Fuel

Reactor Type	Burnup (MWd/MTU)	Initial Activity (Ci/MTU)	1000-year Activity (Ci/MTU)
PWR	33,000 (standard)	3.91×10^5	1.74×10^3
PWR	35,000	1.75×10^5	1.81×10^3
PWR	45,000	2.18×10^5	2.19×10^3
PWR	50,000	2.40×10^5	2.34×10^3
BWR	27,500 (standard)	3.26×10^5	1.58×10^3
BWR	40,000	1.91×10^5	1.94×10^3

Because of the shorter decay time of the standard source terms, the total initial activities for the standard source term (even at lesser burnups) are about a factor of 2 higher than for the detailed source terms. After longer decay, the total activities become closer in value (differing by about 30% for the burnups listed). As is discussed in Chapter 6 of the TSPA-91 report, the greatest releases shown in Figure 2 would occur if a large fraction of the waste package were exhumed at early times. At early times, the differences in activities between the standard and detailed source terms are relatively greater, and the differences in the two curves in Figure 2 at the 10^{-4} probability level (normalized releases greater than 1.0) may be due to this. Lesser releases occur if the inventory has decayed (or less is exhumed). As Table 6 shows, at 1000 years the differences among the activities are also less. In Figure 2, normalized releases in the range 0.1 to 1.0 (with cumulative probabilities less than 0.1) are very similar for the two source terms.

To see the maximum difference possible, Figure 3 compares analyses done assuming the repository inventory is either 1) the base case, 2) entirely composed of the lowest-activity source term (BWR, 10,000 MWd/MTU burnup, 70-year decay), or 3) the highest-activity source term (PWR, 45,000 MWd/MTU burnup, 25-year decay). The base case is considerably closer to the high-activity source than to the

Figure 3
Comparison of Base Case with Highest and Lowest Inventory Activities

lowest activities. Because of the small proportion of low-activity inventory to high-activity inventory for this case, it is not surprising that the results of the two simulations are so close.

Figure 2, which presents releases for the entire inventory, shows that there is not a significant difference in the results between the two source terms for the human-intrusion, surface-release analysis. Since the inventory consists of both long- and short-half-life radionuclides, fission products and actinides, and radionuclides with both straight decay and ingrowth, there may be more pronounced effects for individual radionuclides. Figure 4 shows the radionuclides contributing most to direct surface releases from the standard source term. The values represent means over 10,000 years. Releases by three actinides— ^{240}Pu , ^{241}Am , and ^{239}Pu —are about equal at 30 percent, while ^{238}Pu and ^{137}Cs are the next largest contributors.

Figure 4
Radionuclides Contributing Most to Direct Releases

Figures 5 through 8 show the activities of four of the most important nuclides as a function of burnup and decay. Decay time is shown from 1 year to 10,000 years. Note the small change in inventories as a function of time for ^{240}Pu (Figure 5) and ^{239}Pu (Figure 6), reflecting the long half-lives of these isotopes. Over almost the entire decay period the inventories at a given burnup are essentially constant. Consequently, the use of a fuel age at emplacement of 10 years (i.e., the standard source term) does not make a significant difference. The 6569-year half-life of ^{240}Pu results in the pronounced decrease in inventory at 10,000 years, as seen in Figure 5.

Figure 5
Activity of ^{240}Pu as a Function of Burnup and Decay

Figure 6 shows cuts through the burnup-decay-activity surface for ^{239}Pu at 10-year and 1000-year decay times. Both activities are similar in magnitude and differ by less than a factor of 3 over the burnup range.

Figure 6
Change in Activity as a Function of Burnup for ^{239}Pu at Two Decay Times

Figure 7
Activity of ^{241}Am as a Function of Burnup and Decay
Figure 7 shows the burnup-decay-activity surface for ^{241}Am . This isotope is the daughter of ^{241}Pu (half-life of 14.4 years); because of its longer half-life (432.2 years),

Figure 7 shows the burnup-decay-activity surface for ^{241}Am . This isotope is the daughter of ^{241}Pu (half-life of 14.4 years); because of its longer half-life (432.2 years), there is decay-chain ingrowth. Thus, the initial low inventory of ^{241}Am increases for about 90 years until the ^{241}Pu is exhausted and its own decay becomes apparent. The ^{241}Am inventory varies by over an order of magnitude as burnup changes. Assuming the fuel is older than 10 years at emplacement would result in higher initial inventories of ^{241}Am , because of the decay ingrowth evident in the figure. The standard source term may therefore underestimate early-release inventories for a source-term component that contributes approximately 30% of direct releases. (For releases occurring after a few thousand years the difference is negligible.)

Figure 8 shows the activity surface for ^{137}Cs . Because of its 30-year half-life, there is essentially no inventory after about 300-500 years. Prior to that time, the activity varies by more than one order of magnitude as a function of burnup. Because the standard source term assumes that all the fuel is 10 years old at emplacement, ^{137}Cs inventories out to about 300 years would be overestimated, compared to assuming the fuel is 30-40 years old. This overestimation of the activity is about a factor of 2. The other two nuclides with important releases— ^{238}Pu and ^{90}Sr —will have decay surfaces similar to that of ^{137}Cs , and are not shown.

To summarize, the inventories of individual radionuclides vary between 3 and 20 times as a function of burnup. Currently, total-system performance-assessment analyses attempt to predict releases at an order-of-magnitude resolution because of the other uncertainties in the problem specifications. Thus, a more precise specification of the source term may not improve the accuracy of current analyses. A detailed source-term specification may be required for future total-system analyses or for specialized performance assessments, such as evaluation of the engineered-barrier system. The assumption that the spent fuel is 10 years old at emplacement can lead to an underestimation of the releases for ^{241}Am . A more realistic age of emplacement should be used for future TSPA source terms, especially since it involves no modification of the structure of the inputs to the analyses (i.e., the source term would still consist of one inventory, not 22). An age of 30-40 years would better describe the age of the fuel, if repository loading operations start around the year 2010.

Figure 8
Activity of ^{137}Cs as a Function of Burnup and Decay

4.0 Description of Source Terms for Aqueous Releases

The source terms used for TSPA analyses have “evolved” with each iteration of the performance-assessment process. Four nuclides were used for PACE-90 (Barnard and Dockery, 1991)— ^{237}Np , ^{135}Cs , ^{129}I , and ^{99}Tc . They represented different types of source-release mechanisms, and were considered an adequate representation of that portion of the radionuclide inventory likely to be transported by groundwater. For the TSPA-91 aqueous-release calculations more nuclides were included in the source term, but again, only those radionuclides that could be transported in significant quantities were selected. For nominal aqueous transport, nine radionuclides were used: ^{243}Am , ^{239}Pu , ^{237}Np , ^{234}U , ^{135}Cs , ^{129}I , ^{126}Sn , ^{99}Tc , and ^{79}Se ; for the saturated-zone transport due to human intrusion, ^{241}Am , ^{240}Pu , and ^{14}C were added to the previous nine.

The factors affecting the selection of radionuclides are the amount in the initial inventory and the half-life, solubility and other mobilization processes from the emplaced waste, retardation and colloid formation. If doses are eventually used as TSPA performance measures, then dose effects must also be considered.

The radionuclides used in TSPA-91 were chosen because they represent most of the attributes of the spent-fuel inventory. They include a wide range of half-lives (from 432 years for ^{241}Am to 1.6×10^7 years for ^{129}I). Generally, the radionuclides chosen are those with the greatest inventories in the spent fuel after it has been recently discharged from the reactors; however the ^{237}Np , ^{241}Am , and ^{234}U inventories increase because of decay-chain ingrowth. They are thought to be mobilized from the waste by both matrix alteration and dissolution. These radionuclides include a wide range of solubilities, which is a factor in the timing and magnitude of mobilization from the waste package. Similarly, the chosen radionuclides exhibit a wide range of retardation values, from no retardation (i.e., $K_d = 0$) to essentially total retardation ($K_d = 2000$ ml/g in some units).

The largest contributors to nominal-case aqueous releases predicted in TSPA-91 were ^{99}Tc (~80%), ^{129}I (~19%) and ^{79}Se (~1%). The greatest contributors to doses from nominal-case releases came from ^{99}Tc (~74%) and ^{129}I (~25%). Additionally, ^{237}Np was the largest contributor to doses for the human-intrusion saturated-zone analyses. These calculations were done by Pacific Northwest Laboratories as part of TSPA-91 (Eslinger *et al.*, 1993). There are many factors that can contribute to high doses; these include high radionuclide concentrations (influenced by inventory, retardation, solubility, etc.) and high biologic activity (influenced by pathways and dose effects). These characteristics should be considered when evaluating additional nuclides.

4.1 Evaluation of Individual Radionuclides for Aqueous Source Terms

For the next iteration of the total-system performance-assessment process, the source term is being reexamined to ensure that it effectively measures all the performance criteria. Since the time required for releases due to aqueous transport to reach the accessible environment is considerably greater than for direct surface release, this source term should include radionuclides that have substantial inventories over long periods of time. To evaluate candidates for inclusion in the new source term, the 10,000-year inventory of 33,000-MWd/MTU PWR spent fuel has been examined. Radionuclides present in substantial quantities in this inventory will have been available to be transported by groundwater over the period of performance of the potential repository. Besides these radionuclides, several shorter half-life radionuclides that have significant inventories at 1000 years have also been examined.

To select candidate radionuclides, the 10,000-year inventory has been successively screened. An arbitrary cutoff of the 10,000-year inventory greater than 10^{-4} Ci/MTU was first applied. (Below this value, the radionuclide inventories rapidly drop to the 10^{-7} - 10^{-8} Ci/MTU range, which is probably insignificant.) Isotopes with half-lives shorter than 20 years were excluded; (there is no EPA limit for these radionuclides). Solubilities, sorption factors, and dose conversion factors for the remaining nuclides were then compiled from various sources. The final list is given in Table 7. The table lists both actinides and fission products; the actinides are grouped by their membership in decay chains. The five nuclides with 10,000-year inventories of 0 are some of the most important in the 1000-year inventory. Note that the values listed in the Sorption Factor, Solubilities, and Dose Conversion Factor columns should only be considered indicators; more precise values could depend on a number of specific geochemical and dose-pathway factors. Data were not available for every nuclide property; estimates are shown in these cases.

Table 7
List of Candidate Radionuclides for Aqueous Source Terms

Isotope	10k-year Inventory (Ci/MTU)	Half-life (years)	EPA limit (Ci/MTU)	Sorption K_d Factors ^a (ml/g)	Maximum Solubilities ^b (mole/l)	Dose Conv. Factors ^c (rem/Ci)
^{246}Cm	7.23×10^{-3}	4.73×10^3	0.10	500	10^{-6}	4.5×10^6
^{242}Pu	1.69×10^0	3.87×10^5	0.10	100	10^{-6}	4.1×10^6
^{238}U	3.18×10^{-1}	4.47×10^9	0.10	2	10^{-2}	2.3×10^5
$^{242}\text{Am } m$	0.00	1.52×10^2	0.10	500	10^{-6}	4.2×10^6
^{238}Pu	0.00	8.77×10^1	0.10	100	10^{-6}	3.8×10^6
$^{234}\text{U } \dagger\dagger$	1.99×10^0	2.44×10^5	0.10	2	10^{-2}	2.6×10^5
^{230}Th	1.72×10^{-1}	7.70×10^4	0.01	10	10^{-7}	5.3×10^5
^{226}Ra	1.34×10^{-1}	1.60×10^3	0.10	50	10^{-5}	1.1×10^6
^{210}Pb	1.34×10^{-1}	2.23×10^1	1.00	0	10^{-5}	5.1×10^6
$^{243}\text{Am } \dagger\dagger$	6.68×10^0	7.38×10^3	0.10	500	10^{-6}	4.5×10^6
$^{239}\text{Pu } \dagger\dagger$	2.37×10^2	2.41×10^4	0.10	100	10^{-6}	4.3×10^6
^{235}U	1.99×10^{-2}	7.04×10^8	0.10	2	10^{-2}	2.5×10^5
^{231}Pa	3.58×10^{-3}	3.28×10^4	0.10	0	10^{-5}	1.1×10^7
^{227}Ac	3.58×10^{-3}	2.18×10^1	0.10	250	10^{-6} (e)	1.4×10^7

Table 7, Continued

Isotope	10k-year Inventory (Ci/MTU)	Half-life (years)	EPA limit (Ci/MTU)	Sorption K _d Factors ^a (ml/g)	Solubilities ^b (mole/l)	Dose Conv. Factors ^c (rem/Ci)
²⁴⁵ Cm	6.49x10 ⁻²	8.50x10 ³	0.10	500	10 ⁻⁶	4.5x10 ⁶
²⁴¹ Am †	6.55x10 ⁻²	4.32x10 ²	0.10	500	10 ⁻⁶	4.5x10 ⁶
²³⁷ Np †‡	1.18x10 ⁰	2.14x10 ⁶	0.10	1	10 ⁻²	3.9x10 ⁶
²³³ U	4.81x10 ⁻²	1.58x10 ⁵	0.10	2	10 ⁻²	2.7x10 ⁵
²²⁹ Th	1.65x10 ⁻²	7.34x10 ³	0.10	10	10 ⁻⁷	3.5x10 ⁶
²⁴⁰ Pu †	1.84x10 ²	6.54x10 ³	0.10	100	10 ⁻⁶	4.3x10 ⁶
²³⁶ U	3.53x10 ⁻¹	2.34x10 ⁷	0.10	2	10 ⁻²	2.5x10 ⁵
¹⁵¹ Sm	0.00	9.00x10 ¹	1.00	250	10 ⁻⁸	3.4x10 ²
¹³⁵ Cs †‡	3.44x10 ⁻¹	2.30x10 ⁶	1.00	200	large	7.1x10 ³
¹²⁹ I †‡	3.15x10 ⁻²	1.57x10 ⁷	0.10	0	large	2.8x10 ⁵
¹²⁶ Sn †‡	7.24x10 ⁻¹	1.00x10 ⁵	1.00	100	10 ⁻⁷	1.7x10 ⁴
¹⁰⁷ Pd	1.12x10 ⁻¹	6.50x10 ⁶	1.00	0	10 ⁻⁶	1.4x10 ²
⁹⁹ Tc †‡	1.26x10 ¹	2.13x10 ⁵	10.00	0	large	1.3x10 ³
⁹⁴ Nb	9.11x10 ⁻¹	2.03x10 ⁴	1.00	400	small (e)	5.1x10 ³
⁹³ Zr	1.92x10 ⁰	1.53x10 ⁶	1.00	100	10 ⁻¹⁰	1.6x10 ³
⁹³ Mo	3.58x10 ⁻³	3.50x10 ³	1.00	0	10 ⁻⁶ (e)	1.3x10 ³
⁹⁰ Sr	0.00	2.91x10 ¹	1.00	4	large	1.4x10 ⁵
⁷⁹ Se †‡	3.67x10 ⁻¹	6.50x10 ⁴	1.00	0	large	8.3x10 ³
⁶³ Ni	0.00	9.20x10 ¹	1.00	5	10 ⁻¹	5.4x10 ²
⁵⁹ Ni	4.72x10 ⁰	8.00x10 ⁴	1.00	5	10 ⁻¹	2.0x10 ²
³⁶ Cl	1.19x10 ⁻²	3.01x10 ⁵	1.00	0	large	3.0x10 ³
¹⁴ C †*	4.63x10 ⁻¹	5.73x10 ³	0.10	0	large	2.1x10 ³

† Isotopes used for the saturated-zone source term in TSPA-91.

‡ Isotopes used for the aqueous-transport source term in TSPA-91.

* Isotope used for the gaseous-transport source term in TSPA-91.

(e) Estimated value.

a Data from McKinley and Hadermann (1984), "conservative" values.

b Data from SKI (1991) and Guzowski *et al.* (1983); values are for bare elements in water—solubility data given in TSPA-91 assume solubility is less due to presence of waste package.

c Data from DOE (1988b); dose conversion factor listed is for ingestion.

The actinides in Table 7 are divided into the $4n+2$ ("uranium"), $4n+3$ ("actinium"), $4n+1$ ("neptunium"), and $4n$ ("thorium") decay series. (The first three elements of the uranium series, ^{246}Cm , ^{242}Pu , and ^{238}U , are a branch of the series that joins at ^{234}U .) The radionuclides listed are only portions of their respective decay series. To properly calculate the quantities of daughter products present, the entire decay chains should be used. However, the initial inventories of isotopes heavier than those listed are small, so it is probably sufficiently accurate to start the series as indicated. In each series there are a few isotopes with relatively large 10,000-year inventories (^{242}Pu and ^{234}U in the uranium series, ^{239}Pu in the actinium series, etc.). Although many elements in each series have high sorption values (K_{ds}), some are only minimally retarded. For these radionuclides to be major contributors to releases (or doses) over 10,000 years, they must be mobilized from the waste package and transported to the accessible environment. Of the four decay series, the portion of the uranium chain from ^{242}Am *m*, through ^{210}Pb has the combination of relatively high inventories and low retardations. In addition, ^{230}Th has the lowest EPA limit established (0.01 Ci/MTU), and both ^{226}Ra and ^{210}Pb have relatively high dose conversion factors for ingestion (an important dose pathway for releases from the potential repository).

The actinium series contains the isotope with the largest 10,000-year inventory (^{239}Pu). Furthermore, its daughters ^{235}U and ^{231}Pa are not strongly sorbed, and both ^{227}Ac and ^{231}Pa have high dose conversion factors. The other important plutonium isotope, ^{240}Pu , also has a high inventory, and it contributed significantly to the releases calculated in TSPA-91.

The fission-product isotopes used in TSPA-91 generally have some combination of large solubilities, low retardations, and large dose effects. Of the ones not included previously, ^{59}Ni has a relatively large solubility and a relatively low retardation; ^{36}Cl has large solubility, low retardation, and moderate dose effects.

4.2 Recommendations for Aqueous-Source-Term Radionuclides

The portion of the uranium series ^{242}Am *m*, ... ^{210}Pb contains radionuclides that should be suitable indicators of actinide releases. The portion of the actinium series ^{243}Am , ... ^{227}Ac contains radionuclides with larger dose effects. For analyses only concerned with releases, the uranium series should be included in the source term; for dose calculations, the actinium series should be added. Additionally, ^{237}Np , and its immediate parent ^{241}Am , should remain in the source term because of their high dose effects and the relatively low retardation of ^{237}Np .

The fission products currently in the source term have a variety of properties that make them typical of the entire spent-fuel inventory, and most should therefore be retained. However ^{126}Sn , with its low solubility and high retardation, may not contribute significantly to releases and could therefore be omitted for those calculations. The relatively high solubilities and low sorption of ^{59}Ni and ^{36}Cl make them candidates to be added, but the remaining fission products probably do not contribute anything not already found among the current choices. The new aqueous source term for release calculations would therefore have 13 or 14 radionuclides—6 in a decay chain (^{242}Am , ^{238}Pu , ^{234}U , ^{230}Th , ^{226}Ra , and ^{210}Pb), three other actinides (^{240}Pu , ^{241}Am and ^{237}Np), and 4 fission products (^{135}Cs , ^{129}I , ^{99}Tc , and ^{79}Se). For some analyses, ^{14}C could be included. For dose calculations the 5-member decay chain ^{243}Am , ^{239}Pu , ^{235}U , ^{231}Pa and ^{227}Ac and the fission products ^{126}Sn , ^{36}Cl , and ^{59}Ni could be added.

Tables 8 and 9 summarize the recommended source terms for release and dose calculations, respectively. The column Mean Life shows the average life expectancies of the nuclides. Nuclides with life expectancies of the order of the 10,000-year repository performance period can be transported to the accessible environment without decaying. Those with shorter mean lifetimes may decay to their daughter products, and it will be the latter that are released. Thus, when considering decay chains, the presence of a highly retarded decay product may not prevent its release if its parents can be readily transported. Radionuclides with parameter values likely to cause significant contributions to releases or doses are shaded in the tables.

The probability distribution functions for solubility listed in Tables 8 and 9 are the outcomes of an elicitation of geochemistry experts from Los Alamos National Laboratory. They represent the consensus values from the geochemists and from the references cited for Table 7. Values are considered to be indicative of far-field solubilities. The experts felt that near-field solubilities, which are influenced by near-field effects (e.g., heat, radiation, competing chemical processes) would be too difficult to predict. Furthermore, TSPA analyses treat the near-field source-term effects as an input to the far-field transport processes. Thus, at the point where radionuclides are input to the far-field transport system, far-field processes obtain.

The notation in the table for solubility distribution functions lists the minimum and maximum values, and where appropriate, the expected value and coefficient of variation. Lastly, the type of distribution is listed. For log-uniform and beta distributions, the exponents of the solubilities are listed. Uniform distributions weight the higher values in the range more; log-uniform distributions weight the lower values

more, and the beta distributions are peaked in the range of values. Solubilities of 1.0 mole/l listed in the tables are considered to be sufficiently large that an exact value is not important. Similarly, minimum solubilities in the range of 10^{-10} or 10^{-11} are considered sufficiently small that further precision in their specification will not affect the PA analyses.

Table 8
Recommended Radionuclides for "Release" Source Terms

Isotope	Mean Life (years)	EPA limit (Ci/MTU)	"Indicator" K_d values (ml/g)	Solubility Distribution Functions (mole/l) ^a	Dose Conversion Factor (REM/Ci)
$^{242}\text{Am } m$	219	0.10	1000	10^{-10} , 10^{-6} , -, -, uniform	4.2×10^6
^{238}Pu	126	0.10	100	10^{-10} , 10^{-6} , -, -, uniform	3.8×10^6
^{234}U	352,000	0.10	2	-8, -2, -4.5, 0.20, beta (log)	2.6×10^5
^{230}Th	111,000	0.01	1000	-10, -7, -, -, log uniform	5.3×10^5
^{226}Ra	2300	0.10	200	-9, -5, -7, 0.10, beta (log)	1.1×10^6
^{210}Pb	32	1.00	0	-8, -5, -6.5, 0.08, beta (log)	5.1×10^6
^{241}Am	623	0.10	500	10^{-10} , 10^{-6} , -, -, uniform	4.5×10^6
^{237}Np	3,087,000	0.10	1	-8, -2, -4, 0.20, beta (log)	3.9×10^6
^{240}Pu	9400	0.10	100	10^{-10} , 10^{-6} , -, -, uniform	4.3×10^6
^{135}Cs	3,318,000	1.00	200	1.0	7.1×10^3
^{129}I	22,652,000	0.10	0	1.0	2.8×10^5
^{99}Tc	307,000	10.00	0	1.0	1.3×10^3
^{79}Se	94,000	1.00	0	1.0	8.3×10^3
^{14}C	8300	0.10	0	1.0	2.1×10^3

^a Listed are minimum value, maximum value, expected value, and coefficient of variation (if appropriate), and type of distribution. Data obtained from expert elicitation. Exponents of values are listed for log-uniform and beta distributions.

Table 9
Recommended Radionuclides for "Dose" Source Terms

Isotope	Mean Life (years)	EPA limit (Ci/MTU)	"Indicator" K_d values (ml/g)	Solubility Distribution Functions (mole/l) ^a	Dose Conversion Factor (REM/Ci)
²⁴³ Am	10,600	0.10	500	10 ⁻¹⁰ , 10 ⁻⁶ , -, -, uniform	4.5x10 ⁶
²³⁹ Pu	34,800	0.10	100	10 ⁻¹⁰ , 10 ⁻⁶ , -, -, uniform	4.3x10 ⁶
²³⁵ U	1x10 ⁹	0.10	2	-8, -2, -4.5, 0.20, beta (log)	2.5x10 ⁵
²³¹ Pa	47,300	0.10	1	-10, -5, -, -, log uniform	1.1x10 ⁷
²²⁷ Ac	31	0.10	1000	10 ⁻¹⁰ , 10 ⁻⁶ , -, -, uniform	1.4x10 ⁷
¹²⁶ Sn	144,000	1.00	100	10 ⁻¹¹ , 10 ⁻⁷ , -, -, uniform	1.7x10 ⁴
⁵⁹ Ni	115,000	1.00	5	-6, -1, -2.75, 0.25, beta (log)	2.0x10 ²
³⁶ Cl	434,000	1.00	0	1.0	3.0x10 ³

^a Listed are minimum value, maximum value, expected value, and coefficient of variation (if appropriate), and type of distribution. Data obtained from expert elicitation. Exponents of values are listed for log-uniform and beta distributions.

Almost all of the candidate radionuclides listed in the tables above are the same as those deemed important by other reviews (Kerrisk, 1985; Oversby, 1987). In Kerrisk, the criteria for importance included inventory, EPA release limits, sorption and solubility. The actinides Am, Np, Pu, and U are identified as those for which solubility and sorption data are urgently needed. This observation remains true today. Other important elements identified by Kerrisk include C, Cs, I, Ni, Ra, Sn, Sr, Tc, Th, and Zr. All but Sr and Zr are included in the current list. Oversby uses the NRC source-release regulations (10 CFR 60) as criteria to initially identify 17 elements for which solubility and sorption data would be necessary to permit performance-assessment studies. In order of importance to the contribution to the NRC release criterion, the elements are Am, Pu, Th, U, Cm, C, Np, Ra, Ni, I, Cs, Sn, Se, Zr, Nb, Tc, and Pd. By assuming congruent dissolution of the waste form, I, Cs, Se, and Pd are removed from further consideration. Thus, only Cm is not in the current list.

5.0 Summary

The standard and detailed radionuclide source terms produce performance-assessment results that are essentially the same, given the current level of sophistication of the TSPA analyses. Until analyses are done which are considerably more sensitive to the precise amounts of radionuclides present, there is no need to use a more detailed representation of the burnup and decay of the spent fuel. However, the use of a 10-year decay time at emplacement introduces errors that can be easily avoided by using a more realistic initial decay time of 30-40 years.

There are several individual radionuclides that should be used in future aqueous source terms besides the ones used previously. They will make both the release and dose results more sensitive to the source term, and should make the measures of total-system performance for the potential repository more representative. By including most important members of decay chains, the amounts of the radionuclides will remain more accurate over time.

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APPENDIX

Information from the Reference Information Base Used in this Report

This report contains no information from the Reference Information Base.

Candidate Information for the Reference Information Base

This report contains no candidate information for the Reference Information Base.

Candidate Information for the Geographic Nodal Information Study and Evaluation System

This report contains no candidate information for the Geographic Nodal Information Study and Evaluation System.

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