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Evaluation of Decontamination and Decommissioning Alternatives for a Department of Energy
Nuclear Material Processing Facility - 15265

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ABSTRACT

Ten scenarios for SRS Building 235-F were evaluated relative to groundwater protection through use of a 235-F GoldSimTM¹ fate and transport model. The scenarios included two extreme scenarios, a no action scenario, four D&D grouting scenarios, and three D&D inventory removal scenarios. The fate and transport model simulated contaminant release from four 235-F process areas and the 294-2F Sand Filter and fate and transport through the vadose zone, the Upper Three Runs (UTR) aquifer, and the UTR creek. The model was a stochastic model for which 1,000 realizations were run over 100,000 years for each scenario. In association with the seven D&D alternatives only three groundwater standards out of 25 were exceeded, with the combined radium standard showing the greatest exceedance. The standards were only exceeded at the edge of the building after 10,000 years. It was estimated that there was a 90% likelihood that the combined radium standard would be exceeded for all four D&D grouting scenarios and that a 95% inventory removal from process cells 1-5 would result in an essentially zero percent likelihood that any standards would be exceeded.

INTRODUCTION

SRS Building 235-F is a two-story, reinforced-concrete structure approximately 222 feet long, 109 feet wide and 28 feet high that was constructed in the 1950s. Exhaust air from 235-F (containing primarily residual Pu-238 and Np-237) is passed through double HEPA filtration before discharge to the Building 294-2F Sand Filter through an underground tunnel. Exhaust air is drawn through the sand filter by fans located within the Building 292-2F Fan House. The Sand Filter including the tunnel and the Fan House are also reinforced concrete structures. (WSRC 2003a; Rose 2008; and Musall 2012)

Production of special billets (e.g. containing Np-237) within the Actinide Billet Line (ABL) for irradiation in SRS reactors was the first 235-F mission. The next mission was the fabrication of heat sources from Pu-238 oxide powder for space program applications within the Plutonium Experimental Facility (PEF), the Plutonium Fuel Form (PuFF) Facility, and the Old Metallography Lab (OML). Fabrication processes were developed in PEF, large scale fabrication was carried out in the PuFF Facility, and metallographic examinations of the finished product were conducted in the OML. All metallurgical processes within 235-F (including PEF, PuFF, OML and ABL) were shut-down by 1990. The building's most recent mission provided for the receipt, storage (within vaults), and disbursement of plutonium bearing materials in support of SRS and the US DOE complex. Around October 2006, the vaults were de-inventoried

¹ GoldSimTM is a registered trademark of GoldSim Technology Group LLC.

and the facility was transitioned to a reduced surveillance and maintenance state. (WSRC 2003a; Rose 2008; and Musall 2012)

In-Situ Disposal (ISD) alternatives are under consideration for D&D of 235-F and the 294-2F Sand Filter and are being evaluated in regard to groundwater protection, public/industrial worker protection, and cost. An evaluation of D&D alternative scenarios relative to groundwater protection has been conducted through development and use of a 235-F GoldSimTM fate and transport model. Additionally two extreme scenarios (scenarios 1 and 10 below) that are not D&D alternatives were developed for comparative purposes only. The following scenarios have been evaluated using this model:

- Scenario 1, Generic (inventory on ground surface),
- Scenario 2, No action,
- Scenario 3, Grout PuFF Facility Cells 1-5,
- Scenario 4, Grout PuFF Facility Cells 6-9,
- Scenario 5, Grout 235-F first floor,
- Scenario 6, Grout entire 235-F,
- Scenario 7, 60% inventory removal from PuFF Facility Cells 1-5,
- Scenario 8, 75% inventory removal from PuFF Facility Cells 1-5,
- Scenario 9, 95% inventory removal from PuFF Facility Cells 1-5, and
- Scenario 10, No action with extreme infiltration.

The protectiveness of each of the above ISD scenarios has been evaluated against the following standards (the first seven standards are CERCLA groundwater standards applied at SRS and the eighth is the all-pathways standard from DOE Order 435.1, Radioactive Waste Management):

- Beta-gamma maximum contaminant limit (MCL) of 0.04 mSv/yr (4 mrem/yr);
- Combined Radium (Ra-226 and Ra-228) MCL of 0.185 Bq/L (5 pCi/L);
- Adjusted combined gross alpha MCL of 0.555 Bq/L (15 pCi/L);
- Uranium MCL of 30 µg/L, which results in individual isotope limits of 0.37 (10), 0.0174 (0.47), and 0.37 (10) Bq/L (pCi/L) for U-234, U-235, and U-238, respectively;
- Elemental lead MCL of 15 µg/L;
- Polychlorinated biphenyl (PCB) MCL of 0.5 µg/L;
- Preliminary Remediation Goals (PRGs) for Ac-227, Ac-228, Bi-210, Bi-212, Bi-213, Bi-214, Fr-223, Pb-209, Pb-210, Pb-211, Pb-212, Pb-214, Pu-241, Ra-225, Th-231, and Th-234; and
- The DOE Order 435.1 all-pathways 0.25 mSv/yr (25 mrem/yr) dose limit.

DESCRIPTION OF INPUT DATA

Fig. 1 provides the 235-F fate and transport conceptual model along with a brief description of the input data and the location of the groundwater standard assessment points (CERCLA and DOE Order 435.1).

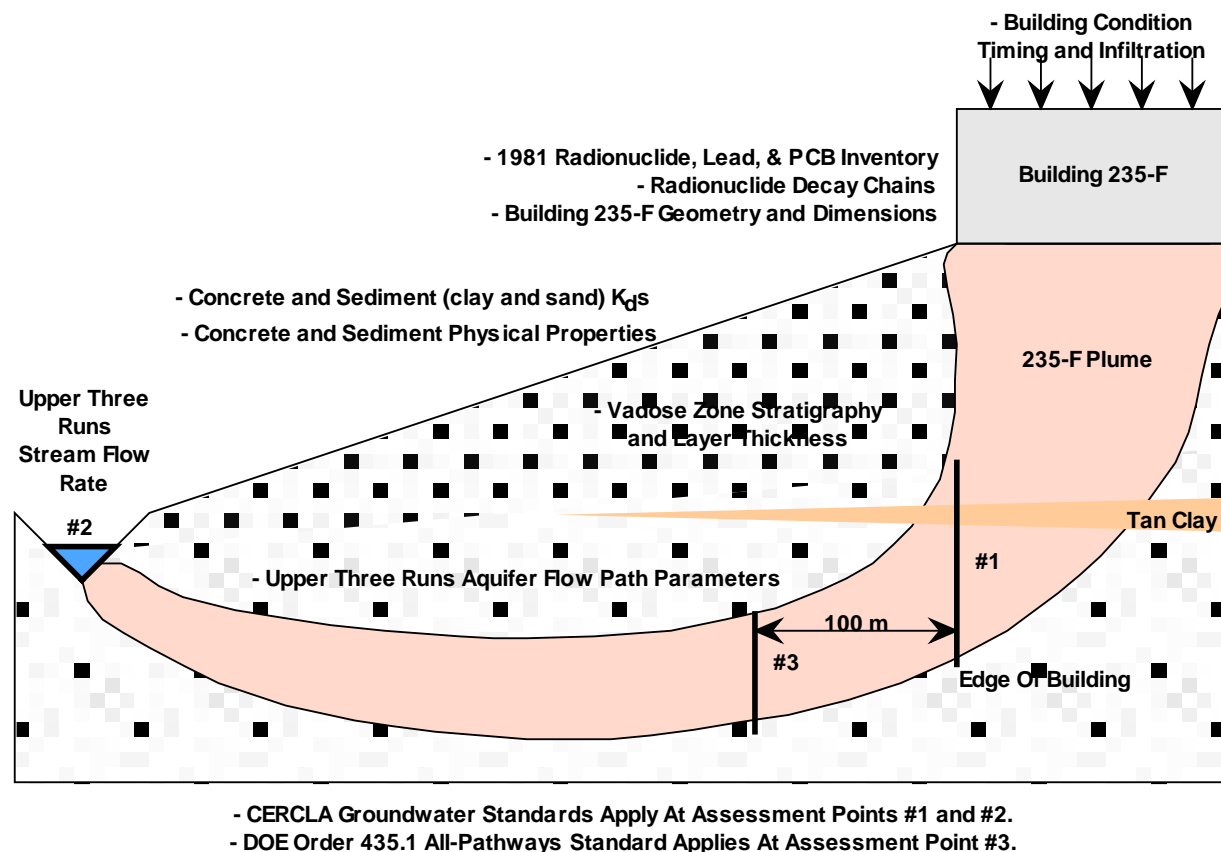


Fig. 1. 235-F fate and transport conceptual model.

Extensive assays of 235-F have been performed and the Table I radionuclide inventory has been developed. As seen, the primary radionuclides are Pu-238, Pu-239, and Np-237 located within PuFF and ABL. In addition to the radionuclides a significant quantity of elemental lead and PCBs are also present in 235-F and have been included in the evaluation. (WSRC 2003a; Rose 2008; and Musall 2012)

Table II presents the ten ISD scenarios and two extreme scenarios evaluated and provides information on the grouting, inventory reduction, and infiltration assumed for each scenario. The grouting considered ranged from no grouting to grouting the entire 235-F building. Inventory reductions considered the reduction of the Table I PuFF Process Cells 1-5 inventory by 60%, 75%, and 95%. Inventory reductions of other portions of 235-F were not considered because of their relatively low inventories compared to that of PuFF Process Cells 1-5. The assumed timing associated with various stages of 235-F collapse was based upon the long-term structural degradation predictions that had been made for similar reinforced concrete structures at SRS (Carey 2005, Carey 2008, and Carey 2009). The infiltration estimates associated with the various stages of 235-F collapse were developed consistent with those developed for SRS Performance Assessments (PAs) and Risk Assessments (RAs) (Phifer et al. 2007, Council 2008, Shine 2008, Council 2009, Taylor and Phifer 2012).

Radionuclide decay chains associated with the Table I radionuclides were extracted from ICRP Publication 107 (ICRP 2008). Only decay modes with branching fractions greater than 1% were included in the calculations. Parents and radionuclide daughters with half-lives greater than three years were explicitly modeled. Daughters with half-lives less than three years were implicitly modeled by assuming secular equilibrium with the closest preceding member in their decay chain for which an activity concentration was calculated.

Other input data (mean and distribution) utilized within the model included the following as described by Taylor and Phifer 2012:

- Building 235-F geometry and dimensions (i.e. building and facility footprints and concrete slab thicknesses),
- Vadose zone stratigraphy and layer thickness (i.e. vadose zone, clayey sediment, and sandy sediment thicknesses)
- Upper Three Runs aquifer flow path parameters (i.e. flow lengths, pore velocity, and travel time through sand and clay and 235-F plume cross-section extracted from a 3-dimensional aquifer model),
- Concrete and sediment distribution coefficients (K_{ds}),
- Concrete and sediment physical properties (i.e. porosity, dry bulk density, particle density, saturation, and tortuosity), and
- Upper Three Runs stream flow rate.

TABLE I. 1981 radionuclide, lead, and PCB inventory

Radionuclide	235-F PuFF Process Cells 1-5 (g)	235-F PuFF Process Cells 6-9 (g)	235-F Actinide Billet Line (g)	Rest of 235-F (g)	235-F Total (g)	294-2F Sand Filter (g)
Pu-238	769.45	2.27	1.17	37.09	809.99	3.01
Pu-239	127.17	0.38	-	6.13	133.67	0.50
Np-237	0.46	1.36E-03	115.97	0.13	116.57	1.80E-03
U-235	-	-	-	32.10	32.10	-
Pu-240	18.43	5.45E-02	-	0.89	19.37	7.20E-02
Pu-241	3.78	1.12E-02	-	0.18	3.97	1.48E-02
Pu-242	1.47	4.36E-03	-	7.11E-02	1.55	5.76E-03
Th-232	0.46	1.36E-03	-	2.22E-02	0.48	1.80E-03
Am-241	0.28	8.17E-04	-	1.33E-02	0.29	1.80E-03
U-233	-	-	1.17E-08	1.11E-11	1.17E-08	-
Th-229	-	-	4.69E-15	4.44E-18	4.69E-15	-
Lead	1.33E+07	3.88E+06	5.16E+06	4.08E+06	2.64E+07	-
PCBs ^a	-	-	-	-	1,195.10	-

^a PCBs are distributed throughout the entire building.

TABLE II. 235-F D&D scenarios (building condition timing and infiltration)

Scenario		Grouting	Inventory Reduction	Infiltration	
#	Name			Building Condition Timing (years)	Infiltration (cm/year)
1	Generic ^a	None	None	>0	38.1 ^a
2	No action	None	None	<350 ^c	1.24 ^g
				>350 ^c	62.5 ^h
3	Grout Cells 1-5	Grout PuFF Facility Cells 1-5	None	<350 ^c	1.24 ^g
				350 to 750 ^d	31.5 ^{i,j}
				>750 ^d	62.5 ^{h,k}
4	Grout Cells 6-9	Grout PuFF Facility Cells 6-9	None	<350 ^c	1.24 ^g
				350 to 750 ^d	31.5 ^{i,j}
				>750 ^d	62.5 ^{h,k}
5	Grout First Floor	Grout 235-F first floor	None	<350 ^c	1.24 ^g
				350 to 750 ^e	31.5 ⁱ
				>750 ^e	62.5 ^h
6	Grout Entire 235-F	Grout entire 235-F	None	<750 ^f	1.24 ^g
				>750 ^f	62.5 ^h
7	Decon 60%	None	60% ^b	<350 ^c	1.24 ^g
				>350 ^c	62.5 ^h
8	Decon 75%	None	75% ^b	<350 ^c	1.24 ^g
				>350 ^c	62.5 ^h
9	Decon 95%	None	95% ^b	<350 ^c	1.24 ^g
				>350 ^c	62.5 ^h
10	Extreme Infiltration	None	None	<350 ^c	1.24 ^g
				>350 ^c	124.8 ^l

^a For the generic scenario the inventory is assumed to be on the ground surface, and the infiltration is assumed to be the mean SRS background infiltration of 38.1 cm/year with a standard deviation of 0.43 cm/year (Shine 2008).

^b PuFF Facility Process Cells 1-5 inventory removal (see Table I).

^c The median timing prior to roof collapse is assumed to be 350 years with a uniform range of 100 to 600 years.

^d The median timing prior to second floor collapse over the grouted PuFF Facility is assumed to be 750 years with a uniform range of 500 to 1,000 years. The second floor collapse over the grouted PuFF Facility is assumed to be constrained from collapse until after roof collapse.

^e The median timing prior to second floor collapse over the grouted 235-F first floor is assumed to be 750 years with a uniform range of 500 to 1,000 years. The second floor collapse over the grouted 235-F first floor is assumed to be constrained from collapse until after roof collapse.

^f The median timing prior to roof collapse over an entirely grouted up 235-F is assumed to be 750 years with a uniform range of 500 to 1,000 years.

^g The infiltration through the intact 235-F concrete roof slab prior to roof collapse is assumed to be 1.24 cm/year with a standard deviation of 0.014 cm/year (Council 2008 and Council 2009).

^h The infiltration through completely collapsed sections of 235-F (i.e. roof and second floor both collapsed) is assumed to be 62.48 cm/year with a standard deviation of 0.71 cm/year (Taylor and Phifer 2012).

ⁱ The infiltration through partially collapsed sections of 235-F (i.e. roof collapsed but second floor intact) is assumed to be 31.5 cm/year with a standard deviation of 0.36 cm/year (Taylor and Phifer 2012).

^j Infiltration over PuFF Facility.

^k Infiltration over rest of 235-F.

^l Infiltration equated to mean SRS annual precipitation of 124.8 cm/year with a standard deviation of 1.4 cm/year (Phifer et al. 2007).

DESCRIPTION OF MODEL

The 235-F GoldSim fate and transport model simulates contaminant release from four 235-F process areas (PuFF Facility cells 1-5, PuFF cells 6-9, ABL, and the rest of the building) and the 294-2F Sand Filter. In addition, it simulates the fate and transport through the vadose zone, the UTR aquifer, and the UTR creek. The model is designed as a stochastic model, and as such it can provide both deterministic and stochastic (probabilistic) results. In general the model results are based on 1,000 realizations over 100,000 years, and the aquifer flow path cross-section emanating from the entire 235-F footprint.

Fig. 2 shows the conceptual structure of the fate and transport model. Each of the sources is treated as a 1-dimensional flow path from the facility (orange) to the vadose zone (olive) to the aquifer (blue). Fig. 2 shows a computational expediency where the “Aquifer” is used to assess the contribution of each individual source to the saturated region and the “Shared Aquifer” is used to combine all source and transport the source term to additional assessment points. There are not flow paths between the “Aquifer” and “Shared Aquifer”, rather equivalent source terms are introduced from the exit cell of the vadose zone to both aquifers.

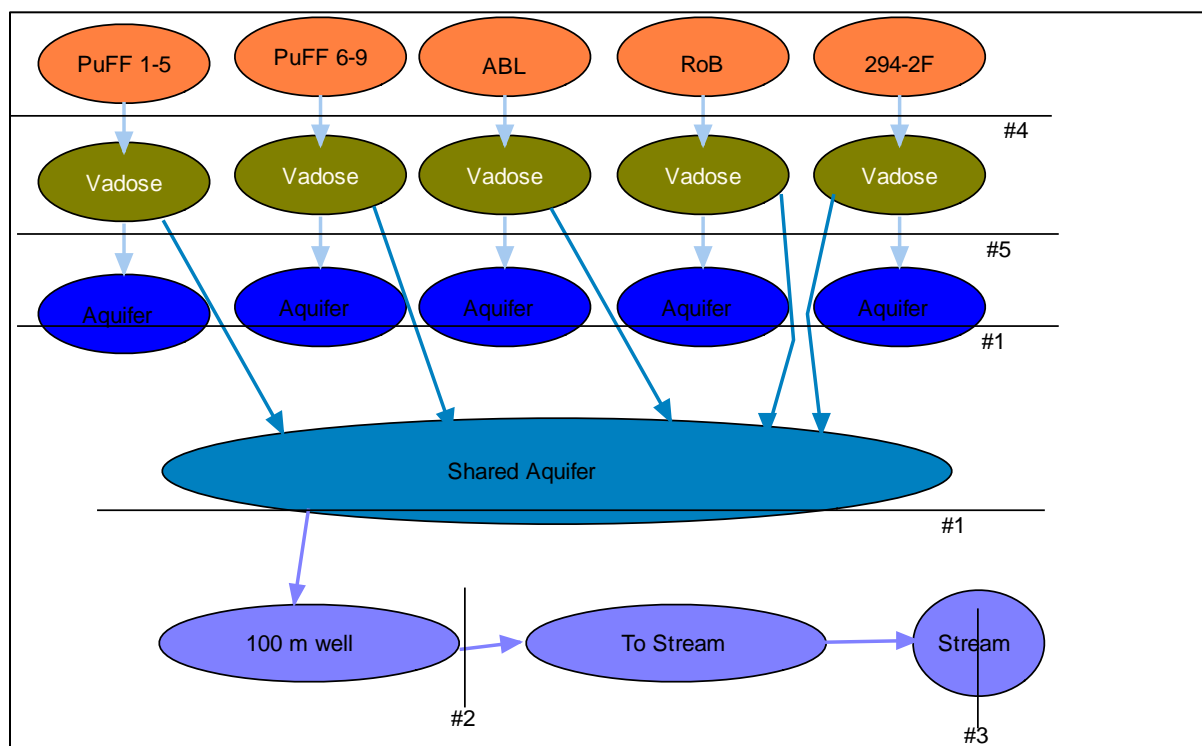


Fig. 2. Conceptual Structure of Computational Model

The model was developed within the GoldSim™ programming environment. GoldSim™ provides the ability to run both deterministic runs, a single realization at specific conditions, and stochastic runs, which consists of a sufficient number of realizations to provide meaningful statistics with input parameters being varied by specified probability distributions. The results of a stochastic run, consisting of multiple realizations, can be used in a sensitivity analysis which can be used to determine to which parameters the model is most sensitive, and, if the model is a reasonable representation of the real world, where the biggest changes can be implemented. The model was designed so that it can be dashboard (Fig. 3) driven. This makes it easy for the user to select various scenarios or combinations of scenarios.

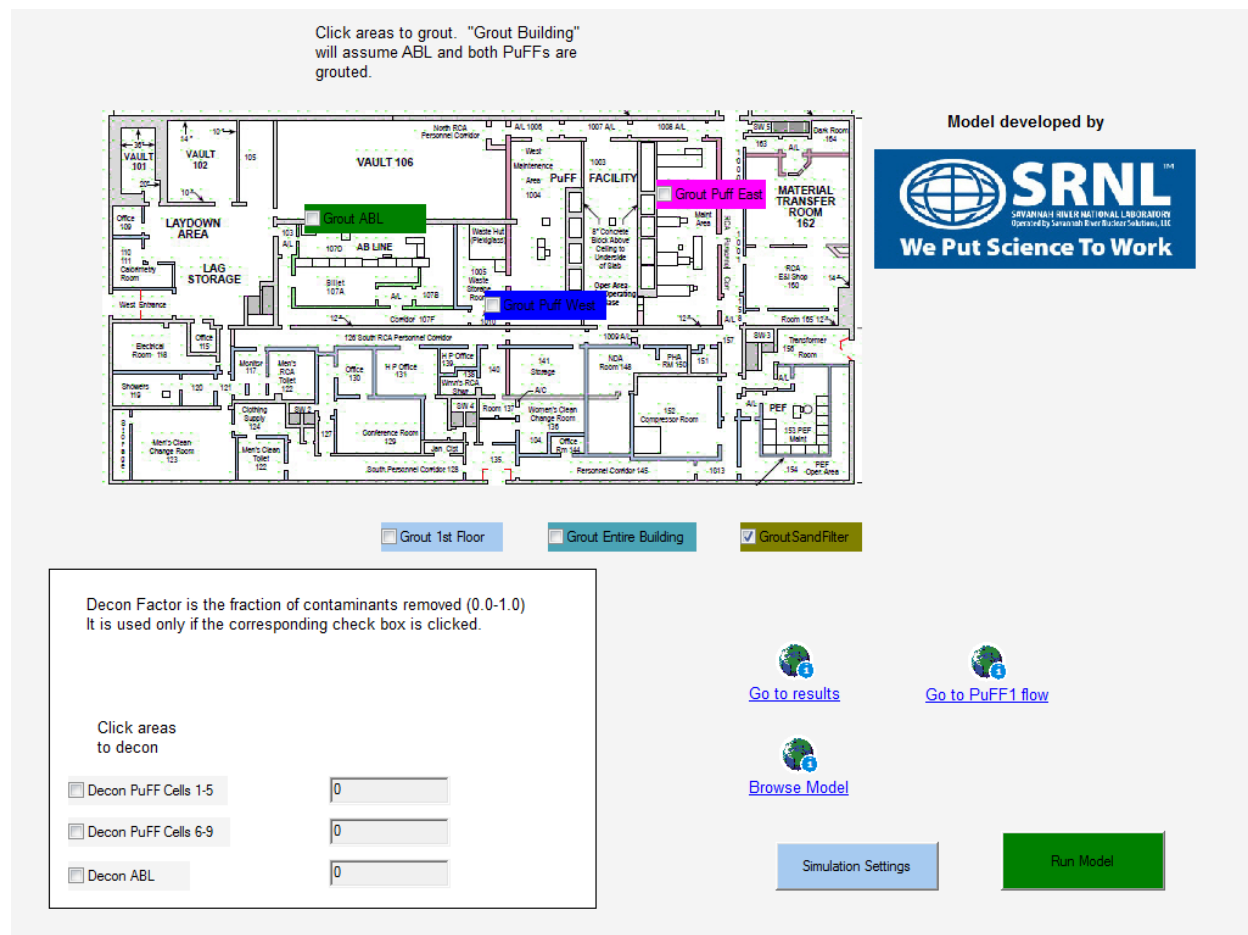


Fig. 3. Model Dashboard

In addition to the transport portion of the model, a dose module is included. The model is based on the E-Area PA and includes updated parameters (both stochastic and deterministic). The following dose pathways considered are based upon the use of well water for domestic purposes, irrigation, and watering livestock:

- Drinking water
 - Ingestion
- Agricultural
 - Ingestion of vegetables

- Ingestion of beef
- Ingestion of milk
- Ingestion of soil
- Inhalation of dust
- External dose from soil

Doses were individually calculated for each source. A combined (all sources) dose was also calculated.

DISCUSSION OF RESULTS

Table III provides the results for the CERCLA groundwater standards or DOE Order 435.1 all-pathways standard which were exceeded by any realization over 100,000 years (results of all scenarios provided for each standard shown). Each standard in Table III has two columns. The first is the fraction of the 1,000 realizations which exceeded the standard (this can be viewed as the probability of exceeding the limit). The second column is the median value of the maximum values of the realizations regardless of when it occurred. Median values which exceed the standard are highlighted.

Table III. Standards exceeded by any realization

Scenario		Combined Radium ^b (Ra-226 and Ra-228) (0.185 Bq/L) (5 pCi/L)		Gross Alpha ^c (0.555 Bq/L) (15 pCi/L)		U-234 (0.37 Bq/L) (10 pCi/L)		Pb-210 PRG (0.0022 Bq/L) (0.06 pCi/L)	
#	Name	Fraction ^c	Median ^d	Fraction ^c	Median ^d	Fraction ^c	Median ^d	Fraction ^c	Median ^d
Assessment Point 1 at Edge of Building (see Fig. 1)									
1	Generic	-	0.910	-	0.477	-	0.41	-	0.0629
2	No action	0.88	0.340	0.2	0.226	0	2.2E-04	0.54	0.0026
3	Grout Cells 1-5	0.9	0.340	0.18	0.226	0	2.2E-04	0.54	0.0026
4	Grout Cells 6-9	0.89	0.340	0.2	0.226	0	2.2E-04	0.56	0.0026
5	Grout first floor	0.9	0.337	0.17	0.155	0	2.2E-04	0.56	0.0026
6	Grout entire 235-F	0.9	0.337	0.17	0.155	0	2.2E-04	0.54	0.0026
7	Decon 60% ^a	0.37	0.159	0.10	0.185	0	1.9E-04	0.27	0.0011
8	Decon 75% ^a	0.14	0.111	0.07	0.170	0	1.9E-04	0.11	0.0009
9	Decon 95% ^a	0	0.048	0.005	0.148	0	1.9E-04	0.0	0.0003
10	Extreme Infiltration	0.64	0.218	0.44	0.488	0	1.5E-04	0.40	0.0018

^a Decon scenarios refer to the amount of contaminant removed from PuFF cells 1-5, with no other action.

^b Limits for each standard are provided in parenthesis below the standard.

^c Fraction of realizations which exceeded the limit. For example, for the "No Action" case 883 realizations out of the 1,000 realizations exceeded the Radium MCL (0.185 Bq/L (5 pCi/L)) resulting in a fraction of 0.88.

^d Median of the maximum values of the realizations (except for the generic scenario, which was performed as a deterministic simulation, where the maximum value is shown). Values highlighted in yellow exceed their standard.

^e Primary contributors to peak are Th-230 (≈90%) and Po-210 (≈10%).

Assessment point 1 (edge of 235-F) is the only location where standards were exceeded. No CERCLA groundwater standards were exceeded at assessment point 2 (Upper Three Runs stream), and the DOE Order 435.1 all-pathways standard was not exceeded at assessment point 3 (100 m from edge of 235-F). Only four standards were exceeded during the first 100,000 years at assessment point 1 (edge of 235-F) in any of the 1,000 realizations, and all four peaked within the 100,000 years. These included combined radium (primarily from Ra-226), gross alpha (primarily from Th-230 and Po-210), U-234, and the Pb-210 PRG. As seen in Table I the parent radionuclide with the greatest inventory is Pu-238, and all of these radionuclides, which resulted in standards being exceed, are members of the Pu-238 decay chain. The generic (inventory on ground surface) and extreme infiltration (infiltration equated to average annual precipitation) scenarios are extreme scenarios that are not D&D alternatives and were developed for comparative purposes only. As seen the generic scenario resulted in the greatest activity concentrations, while the extreme infiltration scenario resulted in generally lower activity concentrations, except for gross alpha. This would indicate that the extreme infiltration scenario resulted in increased dilution. Because the U-234 standard was only exceeded in the generic scenario and not in one of the D&D alternative scenarios, it bears no further discussion.

The following are the primary observations for D&D alternative scenarios that do not involve inventory removal:

- Approximately 90% of the realizations exceed the combined radium standard of 0.185 Bq/L (5 pCi/L) with a median of approximately 0.34 Bq/L.
- Approximately 20% of the realizations exceed the gross alpha standard of 0.555 Bq/L (15 pCi/L) with a median between 0.16 to 0.23 Bq/L.
- Approximately 55% of the realizations exceed the Pb-210 PRG standard of 0.0022 Bq /L (0.06 pCi/L) with a median of 0.0026 pCi/L.

The following are the primary observations for D&D alternative scenarios that involve inventory removal:

- No median exceeds the combined radium, gross alpha, or Pb-210 PRG standards.
- The percentage of realizations exceeding the combined radium, gross alpha, or Pb-210 PRG standards decreases as the amount of inventory removed from PuFF Facility Cells 1-5 increases.
- At a PuFF Facility Cells 1-5 inventory removal of 95%, essentially no realizations exceed the combined radium, gross alpha, or Pb-210 PRG standards.

Because combined radium realizations at assessment point 1 (edge of 235-F) exceed the standard most often further information will focus on combined radium. Table IV provides the combined radium maximum mean concentrations over three time intervals (i.e. mean value of the maximum values of the realizations within each time interval regardless of when it occurred in the time interval). Mean values which exceed the standard are highlighted. This shows that grouting has a slight positive impact during the first 1,000 years compared to no action, but after that it appears to be neutral. The combined radium standard is only exceeded after 10,000 years and only for those scenarios that do not involve inventory removal and only at assessment point

1 (edge of 235-F). The results for the gross alpha and Pb-210 PRG standards showed a similar trend to combined radium with concentrations below standards prior to 10,000 years and greatest after 10,000 years.

Table IV. Combined radium maximum mean concentrations over three time intervals

Combined Radium (Ra-226 and Ra-228) at Assessment Point 1 (Edge of 235-F) (0.185 Bq/L) (5 pCi/L)				
Scenario		0 to 1,000 years (Bq/L)	1,000 to 10,000 years (Bq/L)	10,000 to 100,000 years (Bq/L)
#	Name			
1	Generic	5.55E-06	0.178	0.910
2	No action	2.15E-04	0.152	0.385
3	Grout Cells 1-5	5.92E-05	0.152	0.381
4	Grout Cells 6-9	2.15E-04	0.152	0.385
5	Grout first floor	4.44E-05	0.152	0.381
6	Grout entire 235-F	1.55E-05	0.148	0.377
7	Decon 60%	4.07E-05	0.067	0.174
8	Decon 75%	6.29E-05	0.047	0.122
9	Decon 95%	2.44E-05	0.019	0.052
10	Extreme Infiltration	3.07E-03	0.185	0.248

Fig. 4 shows the combined radium mean concentrations over time at assessment point 1 (edge of 235-F) for the no action scenario. The thick blue line represents the limit. The green line represents the “building’s” contribution, that is, the sum of all sources over the building footprint. The other lines represent each sources’ contribution to the total. The red line represents PuFF cells 1-5, the lavender line represents the rest of the building, the Texas burnt orange represents the sand filter, the lime green line represents PuFF cells 6-9, and the dark purple represents ABL. The plots show a leveling off which implies that the value of the dependent variable has decreased below some maximum value. The major contributor to this exceedance is Ra-226 from Pu-238. The rest of the building and sand filter supply a small fraction. Removing 60% of the PuFF Facility cells 1-5 inventory allows the total mean radium concentration to fall below the 5 pCi/L standard at assessment point 1 (edge of 235-F) as shown in Fig. 5. Fig. 6 shows that the radium limit is exceeded in about 37% of the realizations for the Decon 60% scenario. The results for the gross alpha and Pb-210 PRG standards showed similar trends.

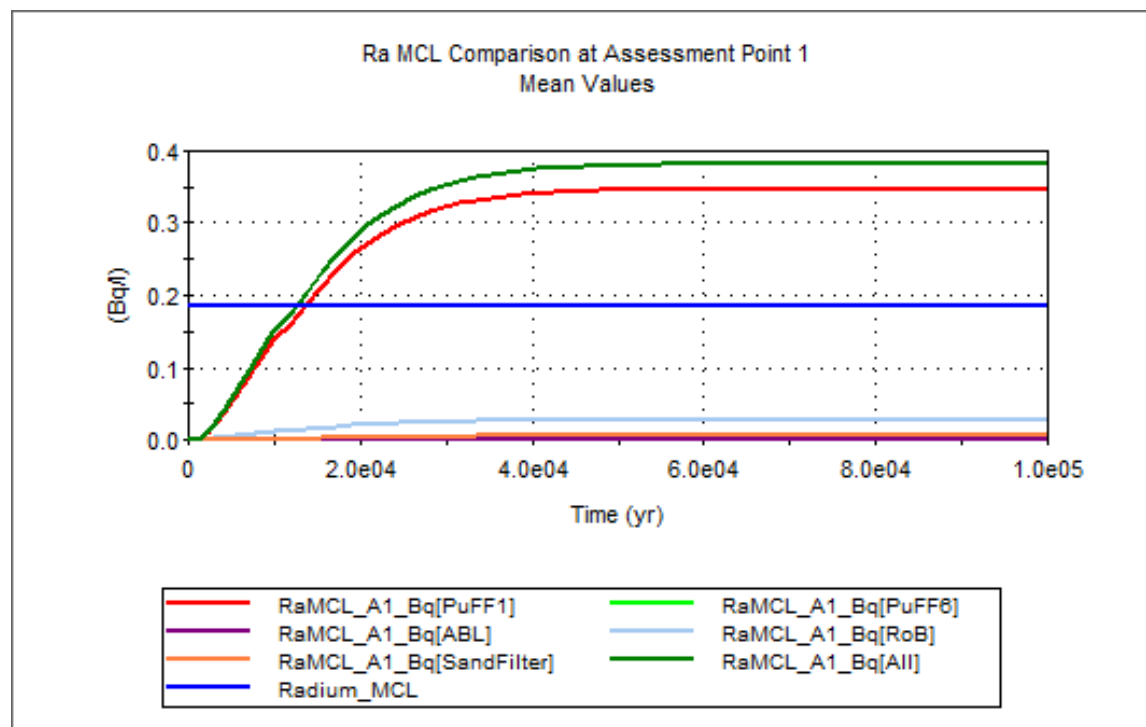


Fig. 4. No action combined radium mean concentrations over time at assessment point 1.

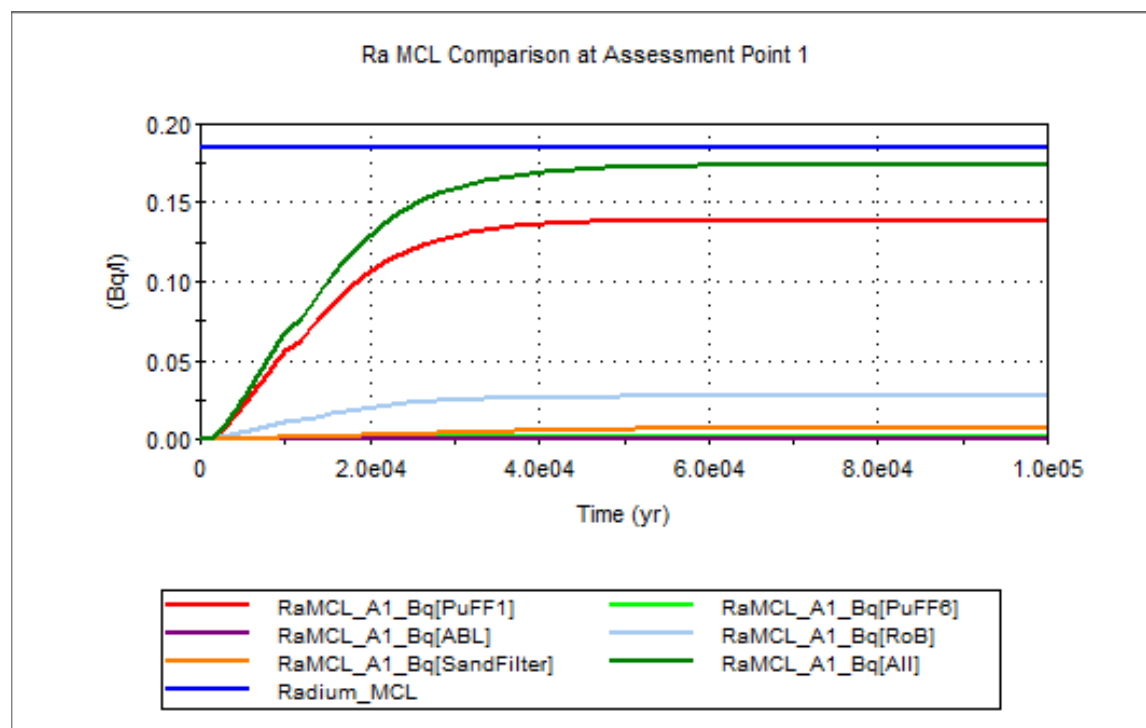


Fig. 5. Decon 60% combined radium mean concentrations over time at assessment point 1.

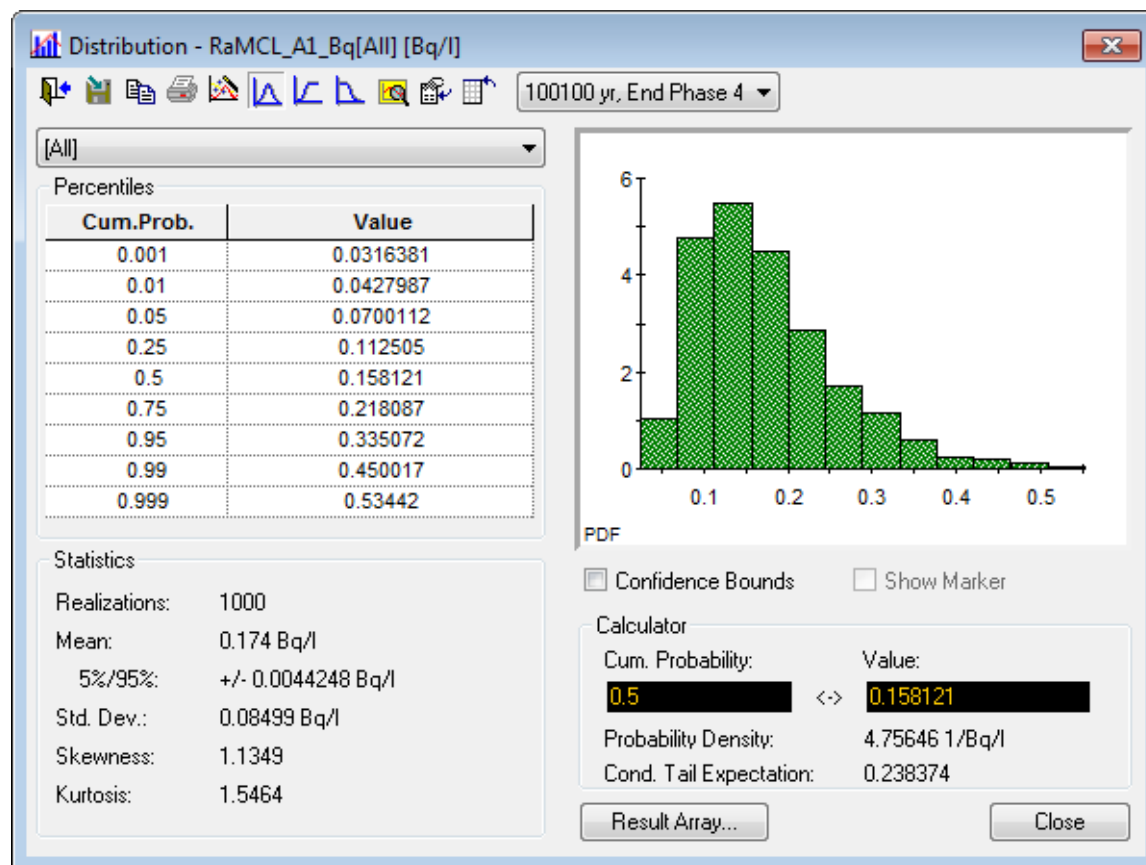


Fig. 6. Decon 60% combined radium mean probability distribution at assessment point 1.

CONCLUSIONS

Of the ten scenarios evaluated only four groundwater standards (combined radium, gross alpha, U-234, and Pb-210 PRG) out of 25 were exceeded for any of the 1,000 realizations over 100,000 years. The U-234 standard was only exceeded in one of the extreme scenarios (i.e. the generic scenario that assumes the entire inventory on the ground surface). Additionally the standards were only exceeded at the edge of the building after 10,000 years. In association with the seven D&D alternatives (i.e. four grouting and three inventory removal scenarios) the greatest level of exceedance was estimated for the combined radium standard followed by the Pb-210 PRG and gross alpha. For the four grouting scenarios the likelihood of exceeding the combined radium, Pb-210 PRG, and gross alpha standards was estimated to be 90%, 55%, and 20%, respectively. The likelihood of exceeding the standards was greatly reduced for the three inventory removal scenarios such that for the 95% inventory removal scenario from PuFF Facility cells 1-5 it was estimated that there was an essentially zero percent likelihood that any standards would be exceeded. It is evident that inventory removal from PuFF Facility cells 1-5 is necessary to avoid exceeding groundwater standards after 10,000 years.

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