Facility: Argonne National Laboratory

Best Practice Title: Using Time Dependent HC Threshold Quantities to Predict the Future Hazard Categorization in DOE Nuclear Facilities

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Brief Description of Best Practice: See attached paper.

Why the best practice was used: In aging < HC-3 nuclear facilities, ingrowth and decay changes the radioactive inventory. We developed a methodology which allows contractors and oversight personnel to quickly determine the effect of ingrowth on their radioactive inventories as a sum of fractions. The methodology uses decay data calculated from the NRC Radiological Toolbox, which uses decay data from ICRP 107, to develop threshold quantities (TQ) that vary with time according to the decay products of the parent. This methodology incorporates the decay products and daughter TQs into a single time dependent TQ that applies to the parent isotope alone and is captured in an Excel Spreadsheet. By doing this, practitioners simply need to know the radiological inventory at a point in time and future inventories in terms of sum of fractions are easily calculated without requiring complex decay calculation and tracking methods.

What are the benefits of the best practice: By creating an Excel spreadsheet file that effectively predicts ingrowth effects to 1000 years, DOE oversight personnel as well as contractors can easily understand the future risk in DOE nuclear facilities using desktop spreadsheet tools.

What problems/issues were associated with the best practice: Very few issues were found. However, we are in the process of applying appropriate software QA to the approach.

How the success of the Best Practice was measured: It is being used at Argonne National Laboratory to understand the future ingrowth and decay inventory of our radiological and nuclear facilities and it has been shared with other SC Laboratories. It was also shared with the DOE ASO for their use in understanding inventories in the New Brunswick Laboratory.

Description of process experience using the Best Practice: The process experience has been very favorable. It is has been used by DOE personnel, Argonne researchers and Argonne operations staff. This work was presented at the 2016 EFCOG Nuclear Facility and Safety Workshop where it was suggested this be submitted as a best practice.

Using Time Dependent Threshold Quantities to Account for Radioactive Progeny and Decay

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Abstract

10CFR830 requires operators of Department of Energy nuclear facilities to appropriately categorize each facility as a Hazard Category 1,2,3 or less than Hazard Category 3 facility (i.e. a radiological facility). The initial categorization is based on the facility's total radioactive inventory and in accordance with DOE-STD-1027-92 or other approved criteria (e.g. NNSA SD G 1027). The inventory is compared against threshold quantity (TQ) criteria in the relevant standard for both a "summation of threshold ratios" as well as a single mass parameter criticality limit. However, accounting for the effects of radioactive progeny and decay can be difficult to implement in radioactive material inventory software due to the complexity of coding the decay chain logic. In these cases, operators may require a method and/or a tool to assure themselves and regulatory authorities that radioactive ingrowth does not threaten the facility hazard categorization. An alternate way to account for the effect of progeny is to derive time dependent threshold functions for each isotope that rely on the calculated activities of the progeny over time and threshold criteria required by regulation. This approach combines the decay physics and the regulatory thresholds into a single time dependent function per isotope thereby simplifying the calculations for practitioners.

The equations for time dependent threshold quantities are derived dependent on progeny calculations and regulatory thresholds. Progeny results for the 1252 isotopes in ICRP 107 were calculated using the "Radiological Toolbox V3.0.0" (http://crpk.ornl.gov/software) at 460 discrete time points with various time intervals out to 1000 years. The results were then combined with regulatory threshold quantities applicable at Argonne National Laboratory into an EXCEL spreadsheet where time dependent TQ values are determined for isotopes of interest in the form of a lookup table. Values between table entries are determined through linear interpolation using native EXCEL functions. The spreadsheet is used to determine the effect of ingrowth and decay given a radioactive material inventory as well as predict future impacts.

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1 BACKGROUND

10CFR830 [1] requires contractors who operate DOE nuclear facilities to appropriately categorize those facilities as either a Hazard Category (HC) 1, 2, 3 or less than Hazard Category 3 facility (i.e. a "radiological facility"). 10CFR830 requires the use of DOE-STD-1027-92 [2] for initial hazard categorization for HC-2, HC-3 and radiological facilities. The initial categorization is based on both the amount and type of isotopes within a facility and must be based on an inventory of all radioactive materials within the nuclear facility. (See Appendix A to Subpart B of 10CFR830). The inventory is compared against threshold criteria in DOE-STD-1027-92 or other approved standards for both a "summation of threshold ratios" (i.e. sum of fractions) as well as a single mass parameter criticality limit. Since some radioactive isotopes decay into progeny isotopes, the regulation requires that the progeny isotopes be included in the contractor's hazard categorization assessment. However, accounting for progeny can be difficult to implement in a radioactive material inventory system due to the complexity of programming radioactive decay chain calculations into inventory software. An alternate way to account for progeny is by deriving time dependent threshold functions for each isotope that rely on the calculated activity of the progeny and threshold criteria required by regulation. Separate effective time dependent threshold quantities can then be derived for both the summation of threshold ratios and criticality limits. This leads to two types of time dependent functions for each parent isotope. The first is an effective threshold quantity for sum of fraction calculations and the second is a time dependent fissile gram equivalent specific activity to determine fissile gram equivalents.

2 DERIVATION OF TIME DEPENDENT THRESHOLD CRITERIA

2.1 Threshold Value for Sum of Fractions Calculation

Consider that a facility's radioactive material inventory is comprised of a total of M items and each item is comprised of I_m different parent isotopes with an item characterization date given by t_m. Each parent has a decay chain associated with a total of J_{mi} isotope progeny. $A_{mij}(t)$ is the activity of the parent's progeny at any time t. $A_{mi0}(t)$ is the activity of the parent i as a function of time in item m. $A_{mi0}(t_m)$ is the initial characterized parent activity with $A_{mij(j\neq 0)}(t_m) = 0$. $A_{mij}(t)$ has units of radioactive activity and is typically expressed in Curies (Ci). The activity for any isotope, including parents and progeny, in the inventory is proportional to the starting activity of the respective parent and is expressed by:

$$A_{mij}(t) = A_{mi0}(t_m) \times P_{mij}(t - t_m)$$

The combination mi serves to identify the item and parent isotope and comes directly from the inventory system. $P_{mij}(t)$ is dimensionless and the result of calculating the activity of the jth progeny isotope using published decay chain information for the parent i with a unit starting activity. A particular parent's $P_j(t)$ is the same for all items since the parents and their decay chains are not dependent on the specific item. Calculating $P_j(t)$ is typically done by using the Bateman's equation formalism [3] and software programs are available to perform these calculations. Following the "Summation of Radionuclide Threshold Ratios" section as noted in DOE-STD-1027-92 page A-2 and letting TQ_{mij} be the progeny isotope's threshold quantity, the sum of fractions ratio from the decay chain for parent i as a function of time ($SOF_{mi}(t)$) in any given item then becomes:

$$SOF_{mi}(t) = \sum_{j=0}^{J_{mi}} \frac{A_{mij}(t)}{TQ_{mij}}$$

Or

$$SOF_{mi}(t) = A_{mi0}(t_m) \sum_{j=0}^{J_{mi}} \frac{P_{mij}(t - t_m)}{TQ_{mij}}$$

A time dependent effective threshold quantity $TQ_{mi}^{Eff}(t)$ can now be defined so that the Sum of Fractions over time is proportional to the starting activity of the parent

$$SOF_{mi}(t) = \frac{A_{mi0}(t_m)}{TQ_{mi}^{Eff}(t)}$$

Where

$$\frac{1}{TQ_{mi}^{Eff}(t)} = \sum_{j=0}^{J_{mi}} \frac{P_{mij}(t - t_m)}{TQ_{mij}} \tag{1}$$

Finally, the sum of fractions for the facility inventory becomes at any time t:

$$SOF(t) = \sum_{m=1}^{M} \sum_{i=1}^{I_m} SOF_{mi}(t) = \sum_{m=1}^{M} \sum_{i=1}^{I_m} A_{mi0}(t_m) \times \frac{1}{TQ_{mi}^{Eff}(t)} \tag{2}$$

The decay chains as well as the threshold quantities are not dependent on which item they are in. Since, the combination mi serves to identify the parent isotope, $TQ_{mi}^{Eff}(t)$ can be determined for all relevant isotopes in the inventory and can then be applied to the appropriate parent isotope in each item. $\frac{1}{TQ_{mi}^{Eff}(t)}$ is then the inverse of the effective threshold value for determining the sum of fractions per unit activity of the parent isotope. Since $\frac{1}{TQ_{mi}^{Eff}(t)}$ tends toward zero as all radioisotopes decay, it is a better choice than $TQ_{mi}^{Eff}(t)$ for performing calculations.

2.2 Threshold Value for Sum of Pu239 Fissile Gram Equivalent

A similar time dependent function can be derived to account for the effect of progeny in fissile gram equivalent calculations. Exhibit A of Argonne National Laboratory's LMS-PROC-45 outlines the way that the Pu-239 Fissile Gram Equivalent ($Pu239\text{-}FGE$) is calculated at Argonne National Laboratory and is necessary in order to ensure that the single parameter mass limit in DOE-STD-1027-92 is not exceeded. The calculation involves the mass of each isotope (m_k) and a dimensionless Pu-239 equivalence factor (PEF_k). The mass is related to the activity of the isotope through the specific activity (SA_k) which normally has the units of Ci/gm of material such that:

$$m_k = \frac{A_k}{SA_k}$$

The PEF_k are listed in LMS-PROC-45, table A.1. The PEF_k is 0 for all isotopes that are not listed in A.1. Equations 1 and 2 of Exhibit A of LMS-PROC-45 provide the total $Pu239\text{-}FGE$ and can be written as:

$$Pu239\text{-}FGE_k = m_k \times PEF_k$$

or

$$Pu239\text{-}FGE_k = A_k \times \frac{PEF_k}{SA_k}$$

with

$$Pu239\text{-}FGE_{tot} = \sum_k Pu239\text{-}FGE_k$$

Again, if we consider that the radioactive inventory is comprised of items each with parent isotopes, we can express the calculation in terms of the fissile gram equivalents within each decay chain and then sum over all of the chains and items. For a decay chain with progeny, the fissile gram equivalent over time for the parent and decay chain in any given item can be written as:

$$Pu239\text{-}FGE_{mi}(t) = \sum_{j=0}^{J_{mi}} \frac{A_{mij}(t) \times PEF_{mij}}{SA_{mij}}$$

Or

$$Pu239\text{-}FGE_{mi}(t) = A_{mi0}(t_m) \sum_{j=0}^{J_{mi}} \frac{P_{mij}(t - t_m) \times PEF_{mij}}{SA_{mij}}$$

Since $Pu239\text{-}FGE_{mi}(t)$ has units of mass and $A_{mi0}(t_m)$and has units of activity, the term in the summation can be thought as the inverse of an isotope dependent specific activity to determine fissile gram equivalent. With this concept, we can rewrite the decay chain contribution to the fissile gram equivalent as:

$$Pu239\text{-}FGE_{mi}(t) = \frac{A_{mi0}(t_m)}{SA_{mi}^{Pu239\text{-}FGE}(t)}$$

Where $SA_{mi}^{Pu239\text{-}FGE}(t)$ is the fissile gram time dependent specific activity for the decay chain of parent i of item m and is given by

$$\frac{1}{SA_{mi}^{Pu239\text{-}FGE}(t)} = \sum_{j=0}^{J_{mi}} \frac{P_{mij}(t - t_m) \times PEF_{mij}}{SA_{mij}} \tag{3}$$

The total fissile gram equivalent then is the sum of all of the parents and items:

$$
\begin{aligned}
Pu239\text{-}FGE_{tot}(t) &= \sum_{m=1}^{M}\sum_{i=1}^{I_m} Pu239\text{-}FGE_{mi}(t) \\
&= \sum_{m=1}^{M}\sum_{i=1}^{I_m} A_{mi0}(t_m) \times \frac{1}{SA_{mi}^{Pu239\text{-}FGE}(t)}
\end{aligned}
\tag{4}
$$

Again, since the decay chains and the specific activities are not item dependent, $\frac{1}{SA_{mi}^{Pu239\text{-}FGE}(t)}$ can be determined for all parent isotopes in the inventory and can then be applied to the appropriate isotopes in the item. This term tends toward zero as the contribution to the fissile gram inventory goes down and is a better choice for performing calculations than $SA_{mi}^{Pu239\text{-}FGE}(t)$.

3 EXAMPLE CALCULATIONS

This section will present how equations 1 through 4 in section 2 can be used to determine $\frac{1}{TQ_{mi}^{Eff}(t)}$ and $\frac{1}{SA_{mi}^{Pu239\text{-}FGE}(t)}$. These time dependent functions can then be used to calculate the effect of progeny on the HC-3 SOF and the Pu-FGE for the radioactive material inventory. Consider a radiological facility that only has a single item ($M = 1$) and that it is comprised of 0.2 Ci of ^{32}Si and 3 Ci of ^{239}Pu. The choice of the isotopes is made for illustration only. One of the ^{239}Pu progeny is ^{235}U. Both this parent and progeny contribute to Pu-FGE in accordance with LMS-PROC-45 [4]. For illustration purposes, the effects of progeny will be determined from 0 to 100 years at 5 year intervals.

3.1 Step 1: Determine the Activity of the Relevant Decay Chains Decay Products

There are various practical approaches for determining the activities in the decay chain of an isotope including manual derivation following Bateman's approach [3] or utilizing a software program that performs these calculations. For this example, decay will be calculated utilizing the Decay Data feature of the Radiological Toolbox Version 3.0.0 written at Oak Ridge National Laboratory for the Nuclear Regulatory Commission [5]. Section 2.3 of the User Guide[6] for the Radiological Toolbox notes that the decay data have been updated to ICRP 107 [7]. One of the advantages of the Radiological Toolbox is that the results can be exported directly to Excel for further processing. While the ^{32}Si has a single radioactive progeny isotope, ^{239}Pu contains 16 progeny isotopes. For clarity, the

^{239}Pu chain will be truncated after the first three progeny to limit the number of columns in the example tables.

3.2 Step 2: Determine the Relevant Physical Constants and Regulatory Thresholds

In addition to the activity of the decay chains, Equation 1 requires knowledge of the threshold quantities for the Hazard Category that is being considered. 10CFR830 [1] requires the use of DOE STD 1027-92 [2] is used for the threshold quantities in this example.. Table 1 shows the activity data along with the HC-3 threshold quantities in the nomenclature of Section 2. The one exception is ^{235m}U in the ^{239}Pu decay chain. In order to calculate the effect of the ^{235m}U, a threshold value must be assumed since there is no clear regulatory basis for a specific value. 20 Ci is assumed for the purpose of this illustration.

In order to calculate the effect of decay chain progeny on the Pu239-FGE, Equation 3 requires the individual specific activities as well as the dimensionless Pu-239 equivalence factors. The equivalence factors are tabulated in LMS-PROC-45 [4] for use at Argonne National Laboratory. The only isotopes in this example that contribute to the Pu-239-FGE are ^{239}Pu and ^{235}U with equivalence factors of 1 and 0.65 respectively. All other factors are zero. Table 2 shows the activity data along with the equivalence factors (PEF_{ijk}) and the specific activities. The specific activities were determined from the summary Decay Data Table in the Radiological Toolbox and then converted to Ci/g.

3.2 Step 3: Calculate the individual terms in Equations 1 and 3.

The calculations of the individual terms for Equations 1 and 3 are in Tables 3 and 4 respectively based on the data in Tables 1 and 2.

3.3 Step 4: Complete the Summations and Calculate the Progeny Effect on the Inventory

Summations are completed for each time point. Row Summation is done for each common Parent column in Tables 3 and 4 to produce tables of $\frac{1}{TQ_{mi}^{Eff}(t)}$ and $\frac{1}{SA_{mi}^{Pu239\text{-}FGE}(t)}$ for use in Equations 2 and 4 respectively. These summations are given in Table 5 for the two parent isotopes in this example. Table 6 multiplies the isotope tables by the assumed inventory activity and further calculates the total HC-3 Sum of Fractions and the Pu239-FGE.

Parent:	Si-32	Si-32	Pu-239	Pu-239	Pu-239	Pu-239
Parent/Progeny:	Si-32	P-32	Pu-239	U-235m	U-235	Th-231
ij	10	11	20	21	22	23
HC3 TQ_{1ij} [Ci]:	52	12	0.52	20*	4.2	55
$t-t_1$[Years]	$P_{110}(t)$	$P_{111}(t)$	$P_{120}(t)$	$P_{121}(t)$	$P_{122}(t)$	$P_{123}(t)$
0	1	0	1	0	0	3.31E-24
5	0.9741	0.9744	0.9999	0.9993	4.92E-09	4.92E-09
10	0.9488	0.9491	0.9997	0.9991	9.84E-09	9.84E-09
15	0.9243	0.9245	0.9996	0.999	1.48E-08	1.48E-08
20	0.9003	0.9006	0.9994	0.9988	1.97E-08	1.97E-08
25	0.877	0.8772	0.9993	0.9987	2.46E-08	2.46E-08
30	0.8542	0.8545	0.9991	0.9985	2.95E-08	2.95E-08
35	0.8321	0.8324	0.999	0.9984	3.44E-08	3.44E-08
40	0.8105	0.8108	0.9989	0.9983	3.94E-08	3.94E-08
45	0.7895	0.7898	0.9987	0.9981	4.43E-08	4.43E-08
50	0.7691	0.7693	0.9986	0.998	4.92E-08	4.92E-08
55	0.7492	0.7494	0.9984	0.9978	5.41E-08	5.41E-08
60	0.7297	0.73	0.9983	0.9977	5.90E-08	5.90E-08
65	0.7108	0.711	0.9981	0.9975	6.39E-08	6.39E-08
70	0.6924	0.6926	0.998	0.9974	6.89E-08	6.89E-08
75	0.6745	0.6747	0.9978	0.9972	7.38E-08	7.38E-08
80	0.657	0.6572	0.9977	0.9971	7.87E-08	7.87E-08
85	0.64	0.6402	0.9976	0.997	8.36E-08	8.36E-08
90	0.6234	0.6236	0.9974	0.9968	8.85E-08	8.85E-08
95	0.6072	0.6074	0.9973	0.9967	9.34E-08	9.34E-08
100	0.5915	0.5917	0.9971	0.9965	9.83E-08	9.83E-08

Table 1: Progeny Activities (P) and HC-3 Threshold (TQ) Values for the Si-32 and the truncated Pu-239 Decay Chains. (See Equation 1). P was determined using the Radiological Toolbox [5]. TQ Values are from Reference [2] and references within [2] when available *: 20 Ci is assumed as a default when no tabulated quantity is available.

Parent:	Si-32	Si-32	Pu-239	Pu-239	Pu-239	Pu-239
Parent/Progeny:	Si-32	P-32	Pu-239	U-235m	U-235	Th-231
ij:	10	11	20	21	22	23
PEF_{1ij}:	0	0	1	0	0.65	0
SA_{1ij} [Ci/g]:	8470	2.86E+05	6.22E-02	3.08E+09	2.16E-06	5.32E+05
$t-t_1$[Years]	$P_{110}(t)$	$P_{111}(t)$	$P_{120}(t)$	$P_{121}(t)$	$P_{122}(t)$	$P_{123}(t)$
0	1	0	1	0	0	3.31E-24
5	0.9741	0.9744	0.9999	0.9993	4.92E-09	4.92E-09
10	0.9488	0.9491	0.9997	0.9991	9.84E-09	9.84E-09
15	0.9243	0.9245	0.9996	0.999	1.48E-08	1.48E-08
20	0.9003	0.9006	0.9994	0.9988	1.97E-08	1.97E-08
25	0.877	0.8772	0.9993	0.9987	2.46E-08	2.46E-08
30	0.8542	0.8545	0.9991	0.9985	2.95E-08	2.95E-08
35	0.8321	0.8324	0.999	0.9984	3.44E-08	3.44E-08
40	0.8105	0.8108	0.9989	0.9983	3.94E-08	3.94E-08
45	0.7895	0.7898	0.9987	0.9981	4.43E-08	4.43E-08
50	0.7691	0.7693	0.9986	0.998	4.92E-08	4.92E-08
55	0.7492	0.7494	0.9984	0.9978	5.41E-08	5.41E-08
60	0.7297	0.73	0.9983	0.9977	5.90E-08	5.90E-08
65	0.7108	0.711	0.9981	0.9975	6.39E-08	6.39E-08
70	0.6924	0.6926	0.998	0.9974	6.89E-08	6.89E-08
75	0.6745	0.6747	0.9978	0.9972	7.38E-08	7.38E-08
80	0.657	0.6572	0.9977	0.9971	7.87E-08	7.87E-08
85	0.64	0.6402	0.9976	0.997	8.36E-08	8.36E-08
90	0.6234	0.6236	0.9974	0.9968	8.85E-08	8.85E-08
95	0.6072	0.6074	0.9973	0.9967	9.34E-08	9.34E-08
100	0.5915	0.5917	0.9971	0.9965	9.83E-08	9.83E-08

Table 2: Progeny Activity (P) , Pu-239 Equivalence Factors (PEF) and Specific Activities (SA) for the Si-32 and the truncated Pu-239 Decay Chains. (See Equation 3). P and SA were determined using Reference 5], PEF was determined from Reference 4.

Parent:	Si-32	Si-32	Pu-239	Pu-239	Pu-239	Pu-239
Parent/Progeny:	Si-32	P-32	Pu-239	U-235m	U-235	Th-231
ij:	10	11	20	21	22	22
$t-t_1$ [Years]	$P_{110}(t)/TQ_{110}$ [1/Ci]	$P_{111}(t)/TQ_{111}$ [1/Ci]	$P_{120}(t)/TQ_{120}$ [1/Ci]	$P_{121}(t)/TQ_{121}$ [1/Ci]	$P_{122}(t)/TQ_{122}$ [1/Ci]	$P_{123}(t)/TQ_{123}$ [1/Ci]
0	1.923E-02	0.000E+00	1.923E+00	0.000E+00	0.000E+00	6.016E-26
5	1.873E-02	8.120E-02	1.923E+00	4.997E-02	1.172E-09	8.942E-11
10	1.825E-02	7.909E-02	1.923E+00	4.996E-02	2.344E-09	1.789E-10
15	1.778E-02	7.704E-02	1.922E+00	4.995E-02	3.517E-09	2.684E-10
20	1.731E-02	7.505E-02	1.922E+00	4.994E-02	4.688E-09	3.578E-10
25	1.687E-02	7.310E-02	1.922E+00	4.994E-02	5.860E-09	4.473E-10
30	1.643E-02	7.121E-02	1.921E+00	4.993E-02	7.029E-09	5.367E-10
35	1.600E-02	6.937E-02	1.921E+00	4.992E-02	8.200E-09	6.262E-10
40	1.559E-02	6.757E-02	1.921E+00	4.992E-02	9.371E-09	7.156E-10
45	1.518E-02	6.582E-02	1.921E+00	4.991E-02	1.054E-08	8.049E-10
50	1.479E-02	6.411E-02	1.920E+00	4.990E-02	1.171E-08	8.944E-10
55	1.441E-02	6.245E-02	1.920E+00	4.989E-02	1.288E-08	9.838E-10
60	1.403E-02	6.083E-02	1.920E+00	4.989E-02	1.405E-08	1.073E-09
65	1.367E-02	5.925E-02	1.919E+00	4.988E-02	1.522E-08	1.162E-09
70	1.332E-02	5.772E-02	1.919E+00	4.987E-02	1.639E-08	1.252E-09
75	1.297E-02	5.623E-02	1.919E+00	4.986E-02	1.756E-08	1.341E-09
80	1.263E-02	5.477E-02	1.919E+00	4.986E-02	1.873E-08	1.430E-09
85	1.231E-02	5.335E-02	1.918E+00	4.985E-02	1.990E-08	1.520E-09
90	1.199E-02	5.197E-02	1.918E+00	4.984E-02	2.107E-08	1.609E-09
95	1.168E-02	5.062E-02	1.918E+00	4.984E-02	2.224E-08	1.698E-09
100	1.138E-02	4.931E-02	1.918E+00	4.983E-02	2.341E-08	1.787E-09

Table 3: Ratio of Parent/Progeny Activity and HC-3 Threshold Values as given in Table 1 (See Equation 1)

Parent:	Si-32	Si-32	Pu-239	Pu-239	Pu-239	Pu-239
Parent/Progeny	Si-32	P-32	Pu-239	U-235m	U-235	Th-231
ij:	10	11	20	21	22	23
$t-t_1$ [Years]	$P_{110}(t)$* PEF_{110}/SA_{110} [gm/Ci]	$P_{111}(t)$* PEF_{111}/SA_{111} [gm/Ci]	$P_{120}(t)$* PEF_{120}/SA_{120} [gm/Ci]	$P_{121}(t)$* PEF_{121}/SA_{121} [gm/Ci]	$P_{122}(t)$* PEF_{122}/SA_{122} [gm/Ci]	$P_{123}(t)$* PEF_{123}/SA_{123} [gm/Ci]
0	0.000E+00	0.000E+00	1.608E+01	0.000E+00	0.000E+00	0.000E+00
5	0.000E+00	0.000E+00	1.608E+01	0.000E+00	1.481E-03	0.000E+00
10	0.000E+00	0.000E+00	1.607E+01	0.000E+00	2.962E-03	0.000E+00
15	0.000E+00	0.000E+00	1.607E+01	0.000E+00	4.445E-03	0.000E+00
20	0.000E+00	0.000E+00	1.607E+01	0.000E+00	5.925E-03	0.000E+00
25	0.000E+00	0.000E+00	1.607E+01	0.000E+00	7.406E-03	0.000E+00
30	0.000E+00	0.000E+00	1.606E+01	0.000E+00	8.883E-03	0.000E+00
35	0.000E+00	0.000E+00	1.606E+01	0.000E+00	1.036E-02	0.000E+00
40	0.000E+00	0.000E+00	1.606E+01	0.000E+00	1.184E-02	0.000E+00
45	0.000E+00	0.000E+00	1.606E+01	0.000E+00	1.333E-02	0.000E+00
50	0.000E+00	0.000E+00	1.605E+01	0.000E+00	1.480E-02	0.000E+00
55	0.000E+00	0.000E+00	1.605E+01	0.000E+00	1.628E-02	0.000E+00
60	0.000E+00	0.000E+00	1.605E+01	0.000E+00	1.776E-02	0.000E+00
65	0.000E+00	0.000E+00	1.605E+01	0.000E+00	1.924E-02	0.000E+00
70	0.000E+00	0.000E+00	1.605E+01	0.000E+00	2.072E-02	0.000E+00
75	0.000E+00	0.000E+00	1.604E+01	0.000E+00	2.220E-02	0.000E+00
80	0.000E+00	0.000E+00	1.604E+01	0.000E+00	2.368E-02	0.000E+00
85	0.000E+00	0.000E+00	1.604E+01	0.000E+00	2.515E-02	0.000E+00
90	0.000E+00	0.000E+00	1.604E+01	0.000E+00	2.663E-02	0.000E+00
95	0.000E+00	0.000E+00	1.603E+01	0.000E+00	2.811E-02	0.000E+00
100	0.000E+00	0.000E+00	1.603E+01	0.000E+00	2.959E-02	0.000E+00

Table 4: Ratio of Parent/Progeny Activity and Specific Activity scaled to the Pu239-FGE Equivalence Factor as given in Table 2. (See Equation 3)

	Time Dependent 1/TQ(t)		Time Dependent 1/SA$^{Pu239\text{-}FGE}$(t)	
Parent:	Si-32	Pu-239	Si-32	Pu-239
i:	1	2	1	2
t-t_1[Years]	1/TQ_{11}(t) [1/Ci]	1/TQ_{12}(t) [1/Ci]	1/SA_{11}(t) [gm/Ci]	1/SA_{12}(t) [gm/Ci]
0	0.019231	1.923077	0	16.08
5	0.099933	1.97285	0	16.08
10	0.097338	1.972455	0	16.08
15	0.094817	1.972258	0	16.08
20	0.092363	1.971863	0	16.07
25	0.089965	1.971666	0	16.07
30	0.087635	1.971271	0	16.07
35	0.085369	1.971074	0	16.07
40	0.083153	1.970877	0	16.07
45	0.080999	1.970482	0	16.07
50	0.078899	1.970285	0	16.07
55	0.076858	1.96989	0	16.07
60	0.074866	1.969693	0	16.07
65	0.072919	1.969298	0	16.07
70	0.071032	1.969101	0	16.07
75	0.069196	1.968706	0	16.06
80	0.067401	1.968509	0	16.06
85	0.065658	1.968312	0	16.06
90	0.063955	1.967917	0	16.06
95	0.062294	1.96772	0	16.06
100	0.060683	1.967325	0	16.06

Table 5: Time Dependent HC-3 Threshold Quantities (1/TQ) and Pu239-FGE Specific Activity for Si-32 and the truncated Pu-239 decay chains from the results in Tables 3 and 4.

	Time Dependent HC-3 SOF			Time Dependent Pu239-FGE [gm]		
Parent/Total:	Si-32	Pu-239	Total	Si-32	Pu-239	Total
Parent Activity (from text)	0.2	3		0.2	3.00	
t-t_1 [Years]						
0	0.003846	5.769	5.773	0	48.23	48.23
5	0.019987	5.919	5.939	0	48.23	48.23
10	0.019468	5.917	5.937	0	48.23	48.23
15	0.018963	5.917	5.936	0	48.23	48.23
20	0.018473	5.916	5.934	0	48.22	48.22
25	0.017993	5.915	5.933	0	48.22	48.22
30	0.017527	5.914	5.931	0	48.21	48.21
35	0.017074	5.913	5.930	0	48.21	48.21
40	0.016631	5.913	5.929	0	48.21	48.21
45	0.0162	5.911	5.928	0	48.21	48.21
50	0.01578	5.911	5.927	0	48.21	48.21
55	0.015372	5.910	5.925	0	48.20	48.20
60	0.014973	5.909	5.924	0	48.20	48.20
65	0.014584	5.908	5.922	0	48.20	48.20
70	0.014206	5.907	5.922	0	48.20	48.20
75	0.013839	5.906	5.920	0	48.19	48.19
80	0.01348	5.906	5.919	0	48.19	48.19
85	0.013132	5.905	5.918	0	48.19	48.19
90	0.012791	5.904	5.917	0	48.19	48.19
95	0.012459	5.903	5.916	0	48.19	48.19
100	0.012137	5.902	5.914	0	48.18	48.18

Table 6: Effect of Progeny on the total HC-3 SOF and the Pu239-FGE for 0.2 Ci of Si-32 and 3 Ci of Pu-239 using Table 5.

4 APPLICATION TO REAL FACILITY INVENTORIES

The equations in Section 2 show that parent specific functions can be calculated from known data such that the effect of progeny on the sum of fractions and the fissile gram equivalent can be reduced to simple time dependent threshold functions for each parent isotope. This approach reduces final implementation complexity by incorporating the decay chain physics calculations and the regulatory limits directly into single time dependent functions per isotope. Creating tables similar to Table 5 then allows the effect of progeny to be calculated with a variety of tools including spreadsheet programs which incorporate lookup and interpolation features. With a finely spaced time grid, linear interpolation between the points becomes practical. A management assurance tool that calculates the effect of progeny can also be built in EXCEL by using the INDEX, MATCH, and FORECAST functions to perform interpolation between points. Since the decay chains are precalculated and used to determine the $\frac{1}{TQ_{mi}^{Eff}(t)}$ and $\frac{1}{SA_{mi}^{Pu239\text{-}FGE}(t)}$ for each parent isotope, the programming challenge to include this approach in radioactive material inventory systems is also significantly reduced. Appropriate care must be taken to determine the accuracy that results from the time intervals that are used to sample the progeny calculations. The 5 year interval taken in Section 3 is only meant as an example of the calculation method and may be insufficient for any detailed work.

In order to construct tables for practical applications as a management assurance tool at Argonne, the Radiological Toolbox [5] was used to determine the decay and progeny profiles for all 1252 isotopes in ICRP 107[7]. The time intervals used were:

0 to 0.099 year at 0.001 year intervals (about every 8.8 hours out to ~37 days),

0.01 to 0.99 year at 0.01 year intervals (about every 3.7 days out to 1 year),

1.0 to 9.9 years at 0.1 year intervals (about every 37 days out to 10 years),

10 to 100 years at 1 year intervals, and,

100 years to 1000 years at 10 year intervals

The Radiological Toolbox [7] has the ability to store output results directly to EXCEL and this feature was used to extract the results from the program. In order to obtain the >500000 individual output calculations, a script to drive the Radiological Toolbox was written in AutoHotkey [8] which is a free and open source scripting language designed to automate repetitive tasks under Microsoft Windows. The progeny results were saved to files and then reordered to the column format similar to the tables in Section 3. EXCEL calculations were used to determine the $\frac{1}{TQ_{mi}^{Eff}(t)}$ and $\frac{1}{SA_{mi}^{Pu239\text{-}FGE}(t)}$ look up tables for each isotope with the HC-3 threshold quantities currently applicable at Argonne . Argonne is currently using threshold quantities for HC-3 and HC-2 based on the methodology in the Supplemental Guidance Document NA-1 SD G 1027 [9].

In order to apply this methodology, practitioners need to be able to calculate the progeny for an isotope's decay chain, and identify the appropriate HC threshold quantities. In some cases, progeny in the decay chains do not have a regulatory driven threshold quantity. While DOE-STD-1027-92 [2] contains default thresholds for HC-2 categorization if an isotope isn't in the tables, there are no regulatory driven defaults for HC-3. For these calculations, 20 Ci was assumed for the HC-3 threshold for the relatively small number of progeny isotopes that were affected in the present Argonne inventory.

5 RESULTS

Once the time dependent $\frac{1}{TQ_{mi}^{Eff}(t)}$ and $\frac{1}{SA_{mi}^{Pu239\text{-}FGE}(t)}$ look up tables are determined, they can be used in a variety of different scenarios provided that the radioactive item inventory system contains an effective characterization date for each item. Argonne's radioactive material inventory system contains these features.. The first scenario to consider is determining the effect of progeny and decay in the current inventory for HC3 SOF. The results for 12 facilities at Argonne are shown in Figure 1. All calculations were performed in EXCEL and apply to the radioactive inventory in the beginning of 2016..

Ignoring the effects of progeny and decay is more conservative than including them in the Figure 1 results. There are a few facilities, where the ingrowth of progeny increases the sum of fractions (see Facility 1 for example). However, in these cases, the increase is small compared to the minimum 0.25 management reserve for HC3 SOF that Argonne requires to ensure that facilities stay below regulatory limits.

The second scenario is in determining the progeny effects over time. Figure 2 shows the progeny and decay effects on the SOF over the next 50 years with the current inventory of the 12 facilities presented in Figure 1. While the SOF in most facilities decays over time, there are facilities (see Facility 1) where the SOF increases due to the effect of radioactive progeny. Being able to project the changes in the SOF into the future allows facility managers to better manage their inventories in order to stay below regulatory thresholds.

In all cases, the effect of progeny ingrowth on quantities related to the single parameter mass limit for criticality for the current inventory in the 12 facilities presented was negligible.

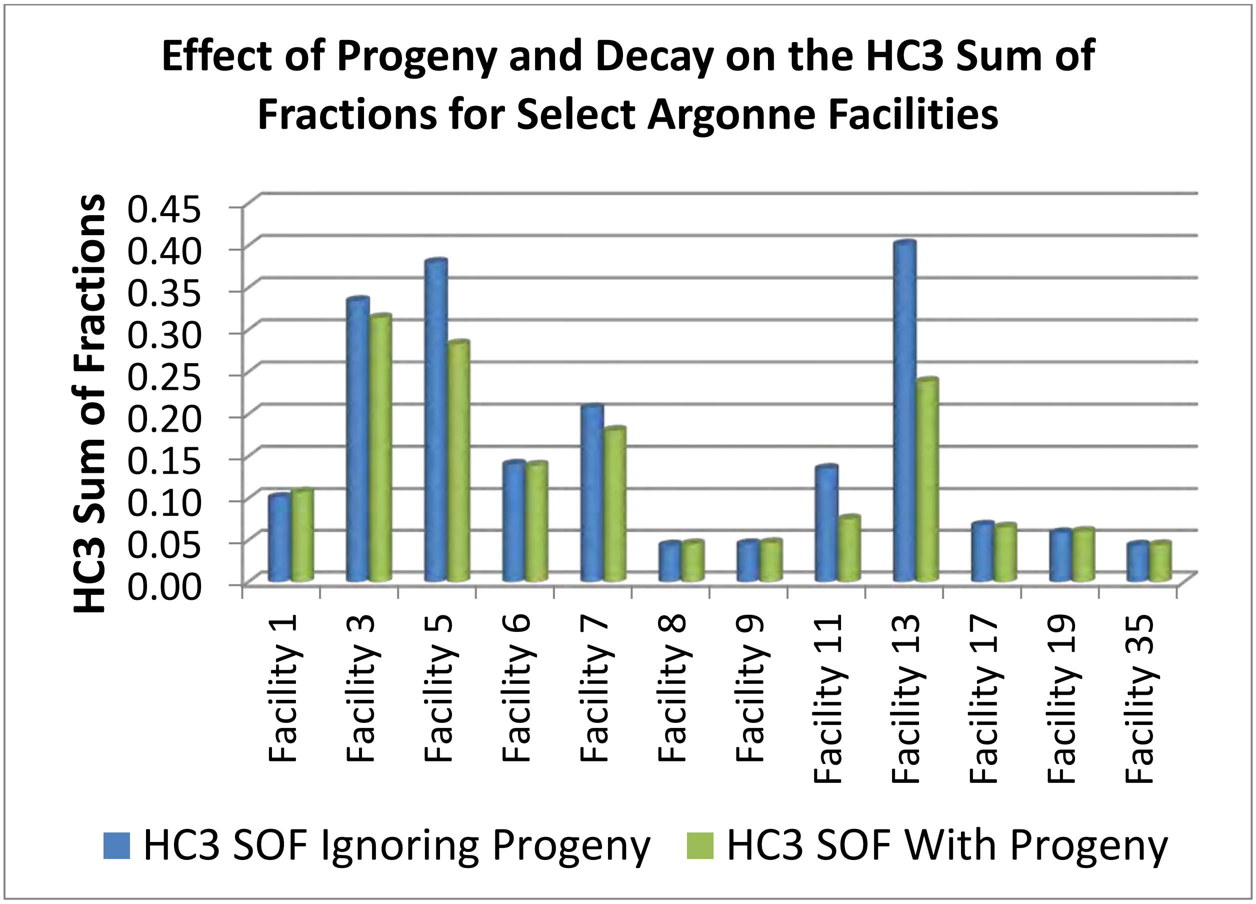

Figure 1: Comparison of including progeny and decay on the SOF for selected Argonne Facilities. HC-2 and HC-3 facilities are excluded.

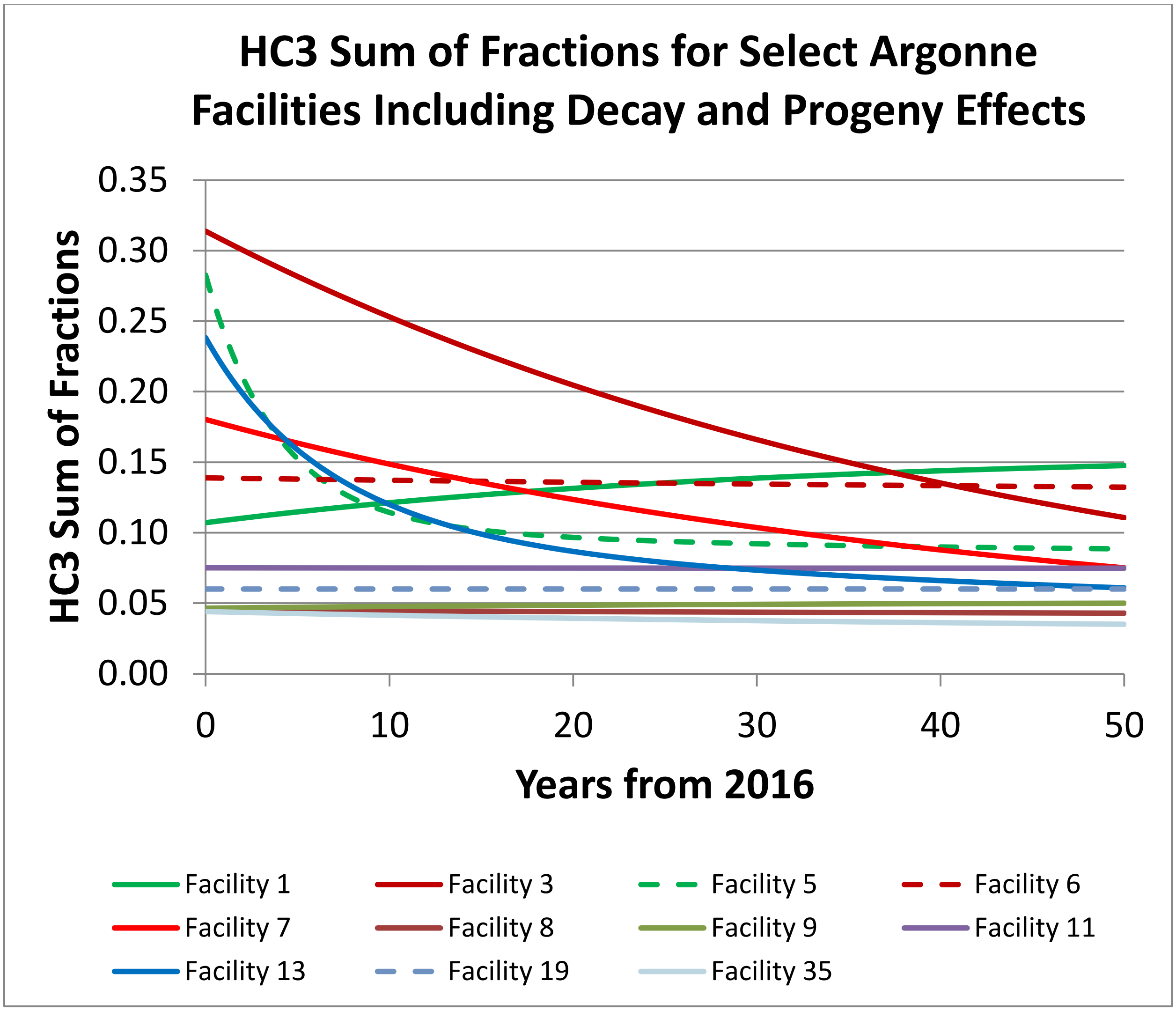

Figure 2: Change in the SOF in selected Argonne Facilities over time. HC-2 and HC-3 facilities are excluded.

6 CONCLUSION

This paper presents a methodology for incorporating radioactive decay and progeny in inventory calculations related to DOE nuclear hazard categorization. The approach derives isotope and time dependent threshold quantities for both sum of fraction calculations as well as fissile gram equivalent calculations based on radioactive decay physics and regulatory thresholds. This approach reduces software implementation complexity by providing a single time dependent function for each isotope.

A description of the approach applicable to spreadsheet calculations is described and results are presented for twelve facilities at Argonne National Laboratory including projecting the impact for the next 50 years. Using this approach, calculations are easily performed in spreadsheet applications such as EXCEL.

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