Effect of Trace Contaminants and Progeny On Hazard Categorizations Assessments

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Outline

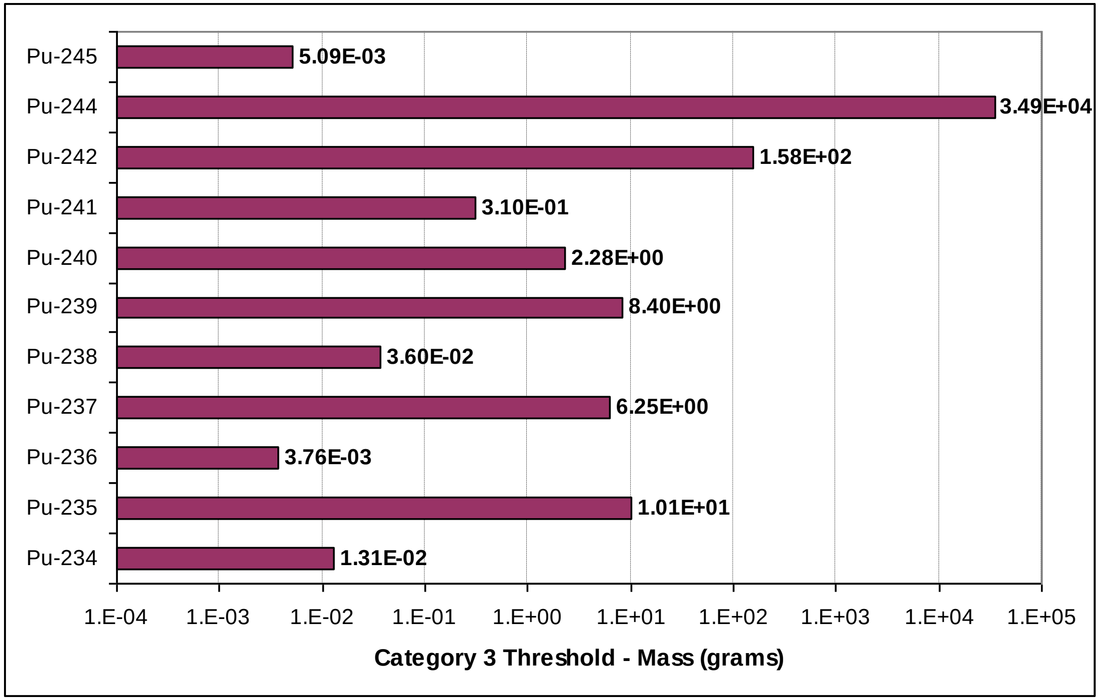
- Contaminants
  - Definition
  - Examples
- Progeny
  - Definition
  - Relationship to HC-3 TQs
  - Examples
- Summary

Contaminants

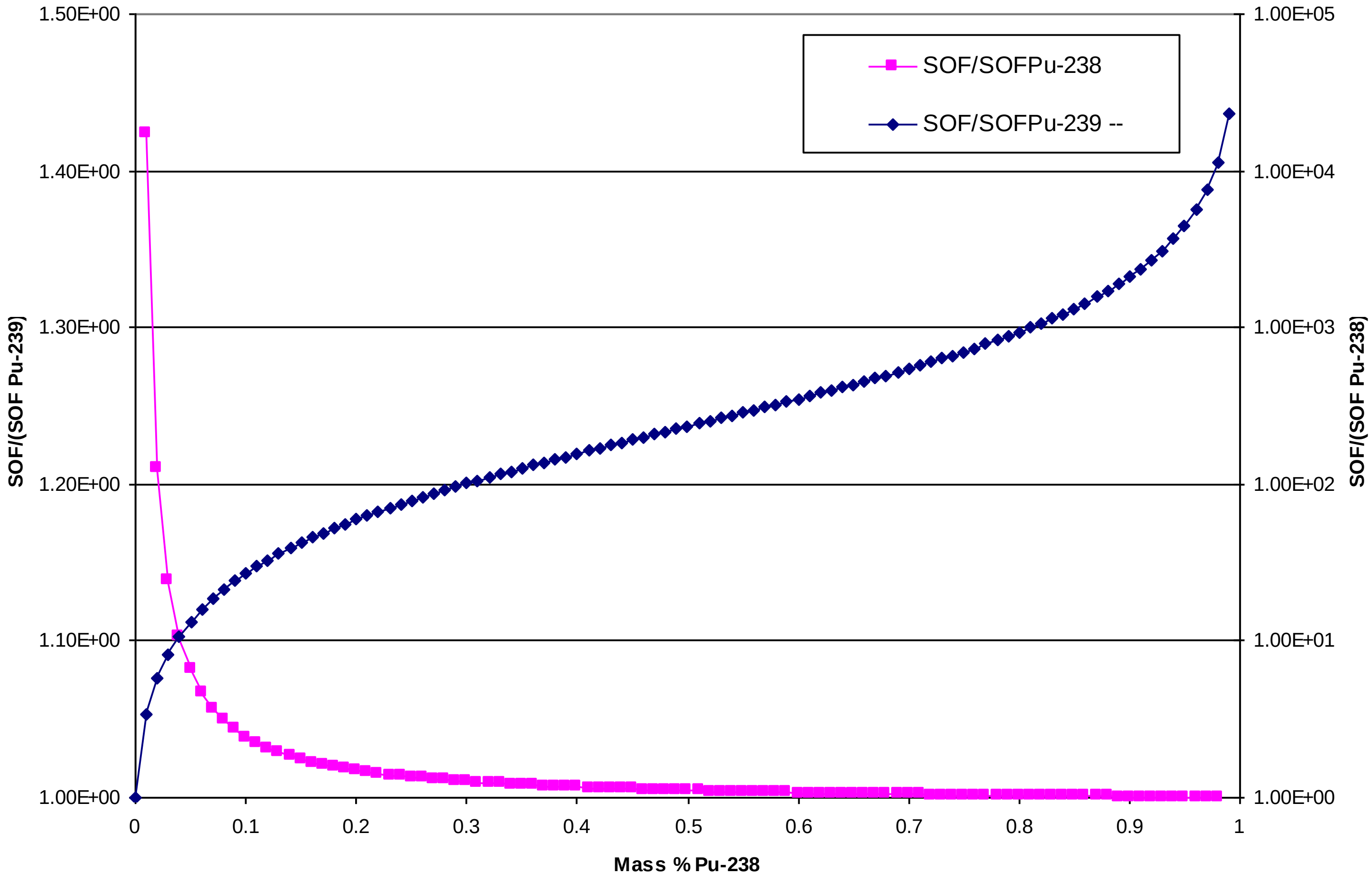
- Definition:
  - Radioactive impurities
  - Usually found in trace amounts in chemically purified radioactive materials.
  - Also present in naturally occurring radioactive materials
  - NOT to be confused with the Health Physics term “Contamination”.
    - This is a radiochemical purity issue.

Contaminants – What's The Big Deal?

- If typically present in trace amounts, then why bother?
  - Because not all TQs (activity / mass) are equal.



^{238}Pu and ^{239}Pu – How each impacts the other as a contaminant



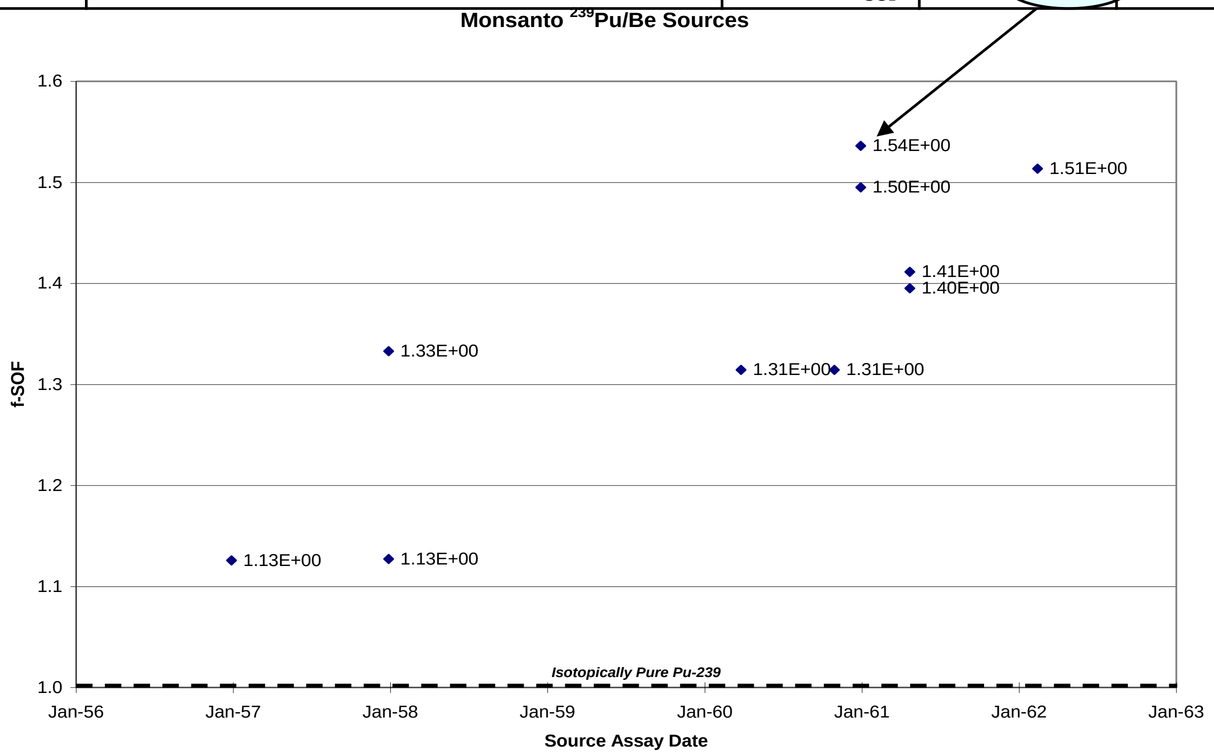
Contaminant Example – Monsanto ^{239}Pu / Be Sources

- Monsanto Research Corporation (MRC) fabricated Pu/Be sources in the 1950's and 1960's
  - Pu/Be sources are used as neutron sources.
  - Paper written in 1968 noted increase in neutron flux from ^{239}Pu/Be sources was due to ^{241}Pu contamination[(1)].
    - ^{241}Pu(β) → ^{241}Am(α)→^{237}Np .....*etc*

(1) M.E. Anderson, "Increases In Neutron Yields of Plutonium-Beryllium (a,n) Sources", Nuclear Applications, Vol. 4, January 1968.

Contaminant Example – Monsanto ^{239}Pu / Be Sources (con't)

Description	Isotope	Atomic Abundance	Activity Abundance	HC-3 TQ (Ci)	SOF / mCi – Pu	% of Total HC-3 SOF
Monsanto ^{239}Pu/Be Neutron Source	^{238}Pu	0.01%	0.21%	6.20E-01	3.42E-06	1.6%
	^{239}Pu	91.6%	7.09%	5.20E-01	1.36E-04	64.9%
	^{240}Pu	7.7%	2.19%	5.20E-01	4.22E-05	20.1%
	^{241}Pu	0.7%	90.50%	3.20E+01	2.83E-05	13.4%
				f_{SOF}	1.54	

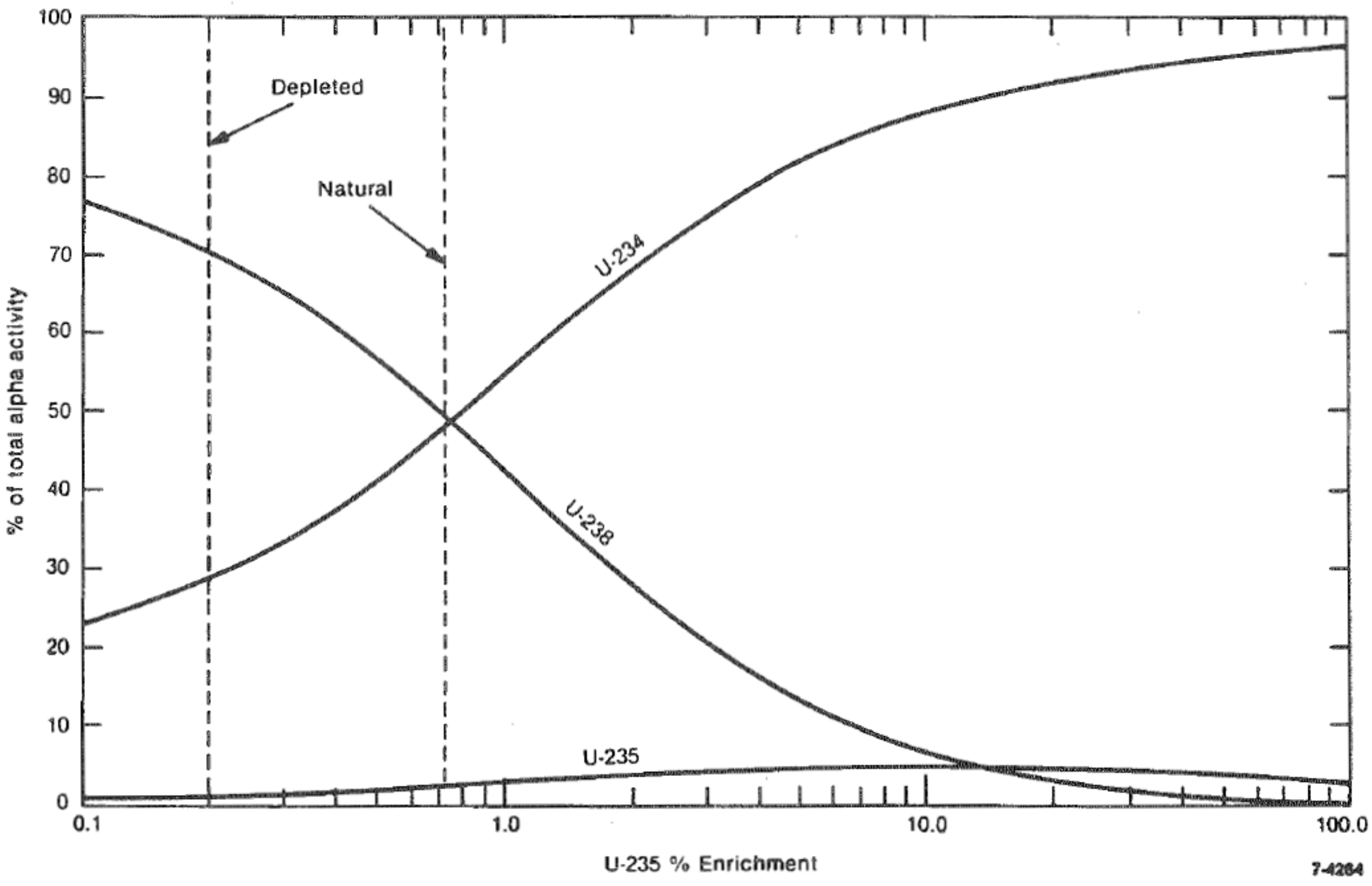


Contaminant Example – Chemically Separated Natural Uranium

- Natural Uranium Radiochemical Purity
  - ~18,000 times more ^{238}U than ^{234}U (by weight).
  - HC-3 TQ for ^{234}U is ~19,000 smaller than the HC-3 TQ for ^{238}U.
    - ^{234}U contribution to HC-3 SOF assessment can be greater than the ^{238}U contribution

Isotope	Wt %	HC-3 TQ (g)	% of Total SOF
^{238}U	99.7823	1.30E+07	48.2%
^{235}U	0.072	1.90E+06	0.2%
^{234}U	0.0055	6.70E+02	51.6%

^{234}U Content In Spectrum of Uranium Source Material



^{234}U Content Haz Cat Effect On Various Uranium Grades

Description	**Isotope**	**Activity Abundance**	**HC-3 TQ (Ci)**	**SOF / Ci – U**
Depleted – Uranium *(0.25% mass ^{235}U)*	^{234}U	8.4%	4.2	2.00E-02
	^{235}U	1.5%	4.2	3.57E-03
	^{238}U	90.1%	4.2	2.15E-01
			$\mathbf{f_{SOF}}$	**1.11**
Natural – Uranium *(0.72% mass ^{235}U)*	^{234}U	48.83%	4.2	1.16E-01
	^{235}U	2.34%	4.2	5.57E-03
	^{238}U	48.83%	4.2	1.16E-01
			$\mathbf{f_{SOF}}$	**2.05**
Enriched (5 wt%) – UF_6 *($10x10^3$ μg $^{234}U/g^{235}U$)*	^{234}U	87.98%	4.2	2.09E-01
	^{235}U	3.04%	4.2	7.24E-03
	^{238}U	8.98%	4.2	2.14E-02
			$\mathbf{f_{SOF}}$	**11.1**
	Isotope	**Mass Abundance**	**HC-3 TQ (Ci)**	**SOF / Ci – U**
Enriched (2 wt%) – UF_6 *($11x10^3$ μg $^{234}U/g^{235}U$)*	^{232}U	1.00E-10	0.82	1.48E-03
	^{234}U	0.02%	4.2	1.85E-01
	^{235}U	2.00%	4.2	5.83E-03
	^{236}U	2.50E-04	4.2	2.18E-03
	^{238}U	97.95%	4.2	4.44E-02
			$\mathbf{f_{SOF}}$	**5.36**

Other Contaminant Examples – ^{241}Am

- ^{241}Am Source Material
  - ^{242m}Am and ^{243}Am are expected contaminants
    - Up to 0.4 wt% ^{242m}Am from LWR[(2)]
    - Minimal HC-3 SOF impact (f_{SOF} = 1.01)
    - Significant ^{235}U FEM contributor

Nuclide	Mass (g)	FEM Factor	FEM (g)
^{241}Am	0.95	0.044	4.18E-02
^{242m}Am	0.004	54	**2.16E-01**
^{243}Am	0.046	0.028	1.29E-03
		TOTAL FEM	**2.59E-01**
		$\mathbf{f_{FEM}}$	6.20E+00

(2) P. Benetti et. al., "Americium 242m and Its Potential Use In Space Applications", Journal of Physics: Conference Series. Vol 41 (2006), pp 161 – 168.

Other Contaminant Examples

Source Material	f_{SOF}	Contaminants Impacting HC-3 SOF	f_{SOF}	Contaminants Impacting ^{235}U FEM
^{252}Cf	1.08	^{250}Cf	--	^{245}Cm, ^{247}Cm, ^{249}Cf, ^{251}Cf
^{248}Cm	1.97	^{246}Cm	--	^{245}Cm, ^{247}Cm
^{244}Cm	1.00	--	8.94	^{245}Cm
^{155}Eu	1.05	^{152}Eu, ^{154}Eu	--	--
^{57}Co	1.56	^{56}Co, ^{58}Co	--	--

Progeny

- Definition:
  - Radioactive isotopes created through the natural radioactive decay of a parent radioisotope.
  - Also referred to as “daughter” isotopes.

What is the Role of Progeny In Haz Cat Assessments?

- HC-3 TQs only includes the dose contribution of progeny that is produced within the body (ICRP-30)
  - ICRP-30 (ALIs) →
  - EPA RQs →
  - DOE HC-3 TQs !
- Progeny produced outside of the body would not be accounted for in the derivation of the ALIs.
  - Progeny that contribute to dose and are present in facility inventory must be addressed in HC-3 Haz Cat assessment.

Methodology for assessing progeny

- Screening criteria
  - 757 Radioisotopes with HC-3 TQs
    - Radioisotopes w/ $T_{1/2}$ > 0.25yr
    - Radioactive progeny
      - 79 radioisotopes left after screening
- Calculation of progeny impact of HC-3 SOF
  - Used Bateman Equations
  - Identified parents that had f_{SOF} factor greater than or equal to 1.10 within 100 years.
    - 37 Radioisotopes
- Two classes of progeny impact
  - Secular equilibrium
    - (SOF increases, then decreases)
  - Non-secular equilibrium
    - (SOF decreases slower than expected)

Progeny In Secular Equilibrium (N, N+1, N+2 and N+3 series)

Isotope	Decay Series	f_{SOF} @ 100 yr	Significant Progeny Contributing to f_{SOF} (% of SOF @ 100 yr in-growth)
^{232}Th	^{232}Th Series (N)	1.16E+00	^{228}Ra (0.7%), **^{228}Th (8.6%)**, **^{220}Rn (4.3%)**
^{228}Th	^{232}Th Series (N)	1.56E+00	^{224}Ra (0.3%), **^{220}Rn (32.2%)**, ^{212}Pb (0.2%), **^{208}Tl (3.2%)**
^{232}U	^{232}Th Series (N)	2.34E+00	**^{228}Th (36.0%)**, ^{224}Ra (0.2%), **^{220}Rn (19.0%)**, ^{212}Pb (0.1%), **^{208}Tl (1.9%)**
^{228}Ra	^{232}Th Series (N)	2.90E+01	**^{228}Th (61.9%)**, ^{224}Ra (0.3%), **^{220}Rn (31.0%)**, ^{212}Pb (0.2%), **^{208}Tl (3.1%)**
^{245}Cm	^{233}U Series (N+1)	1.14E+00	^{245}Pu (1.4%), **^{241}Am (10.5%)**
^{233}U	^{233}U Series (N+1)	1.42E+00	**^{229}Th (29.7%)**, ^{225}Ac (0.1%)
^{210}Pb	^{238}U Series (N+2)	1.19E+00	^{210}Bi (0.1%), **^{210}Po (5.1%)**
^{242m}Am	^{238}U Series (N+2)	1.48E+01	^{242}Cm (0.9%), **^{238}Pu (31.4%)**
^{226}Ra	^{238}U Series (N+2)	**4.06E+01**	^{222}Rn (3.0%), **^{210}Pb (79.4%)**, ^{210}Bi (0.1%), **^{210}Po (15.0%)**
^{235}U	^{235}U Series (N+3)	1.19E+00	^{231}Pa (3.7%), **^{227}Ac (12.4%)**
^{231}Pa	^{235}U Series (N+3)	5.58E+00	**^{227}Ac (81.9%)**, ^{227}Th (0.1%), ^{223}Ra(0.1%)

Significant Progeny Not In Secular Equilibrium

- Parent decays into a longer lived progeny.
  - 17 parents had longer-lived progeny with f_{SOF} >1.10.
    - 11 were transuranium isotopes.
  - Selected examples (where f_{SOF}=1.10):
    - ^{241}Pu ($t_{1/2}$ =14.4 y) → ^{241}Am @ 1.0 year
    - ^{244}Cm ($t_{1/2}$ =18.1 y) → ^{240}Pu @ 77 years
    - ^{250}Cf ($t_{1/2}$ =13.1 y) → ^{246}Cm @ 55 years
- The SOF of mixture typically decreases over time, just not as fast due to progeny build-in.
  - Exception:
    - ^{241}Pu decay into ^{241}Am.
      - Overall SOF increase of 1.86 times (@ 55 y).

Four EPA Defined Mixtures

- The EPA defines mixture RQs for four defined radioactive material mixtures:
  - Natural U (chemical separated)
  - Natural U in secular equil. w/ progeny
  - Natural Thorium
  - ^{226}Ra in secular equil. w/ progeny

An HC-3 Analysis of the Four EPA Defined Mixtures

- Nat U – Chemically Separated
  - ^{238}U, ^{235}U, ^{234}U
  - No progeny
  - f_{SOF} = 2.2
- Natural Thorium
  - ^{232}Th and ^{228}Th
  - f_{SOF} = 1.16
- Nat U w/ progeny
  - Characteristic of uranium ore.
  - ^{238}U, ^{235}U, ^{234}U and associated progeny in sec. equil.
  - f_{SOF} = 29.3
- ^{226}Ra w/ progeny
  - ^{226}Ra and associated progeny in sec. equil.
  - f_{SOF} = 41.9

Contaminants Summary

- Be ready to ask the following:
  - Where did the source come from / who fabricated the source?
    - NORM
    - Reactor (LWR vs. HWR, Commercial vs. High Neutron Flux)
    - Accelerator
  - How was the source material produced?
    - Contaminant composition can be influenced by:
      - Isotopic composition of starting material
      - Irradiation history
      - Chemical purification techniques
  - How old is the source?
    - In-growth issues can impact SOF and FEM assessment
- What to look for:
  - Contaminants with HC-3 SOF TQs smaller than the predominant isotope.
  - Contaminants with ^{235}U FEM factor larger than the predominant isotope.
    - Also applies to ^{239}Pu FGE assessments.

Progeny Summary

- Progeny in secular equilibrium results in higher SOF.
- Progeny in non-secular equilibrium typically has SOF decrease at slower rate.
  - Beware of the exceptions:
  - $^{241}Pu \rightarrow {}^{241}Am$ build-in increases starting HC-3 SOF by 1.86 times (55y).